

# 9-(2-Methoxybenzylidene)-6-(2-methoxyphenyl)-4,8-diphenyl-2-oxa-3,7-diazaspiro[4.4]nona-3,7-dien-1-one

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## Key indicators

Single-crystal X-ray study

$T = 293\text{ K}$

Mean  $\sigma(\text{C}-\text{C}) = 0.003\text{ \AA}$

$R$  factor = 0.041

$wR$  factor = 0.097

Data-to-parameter ratio = 17.2

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

The title compound,  $\text{C}_{33}\text{H}_{26}\text{N}_2\text{O}_4$ , is derived from an unexpected spiran synthesis. Its crystal structure has been determined at room temperature. The skeleton is composed of a spiro[4.4] system formed by an isoxazolone and a substituted pyrroline ring obtained by a series of tandem reactions. The carbonyl exocyclic bond angles [ $121.9(2)$  and  $129.9(2)^\circ$ ] of the isoxazolone ring show the usual asymmetry.

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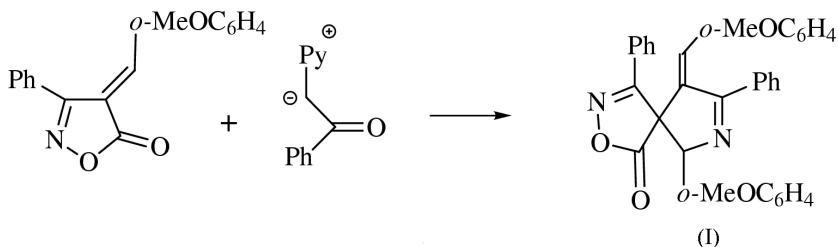
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## Comment

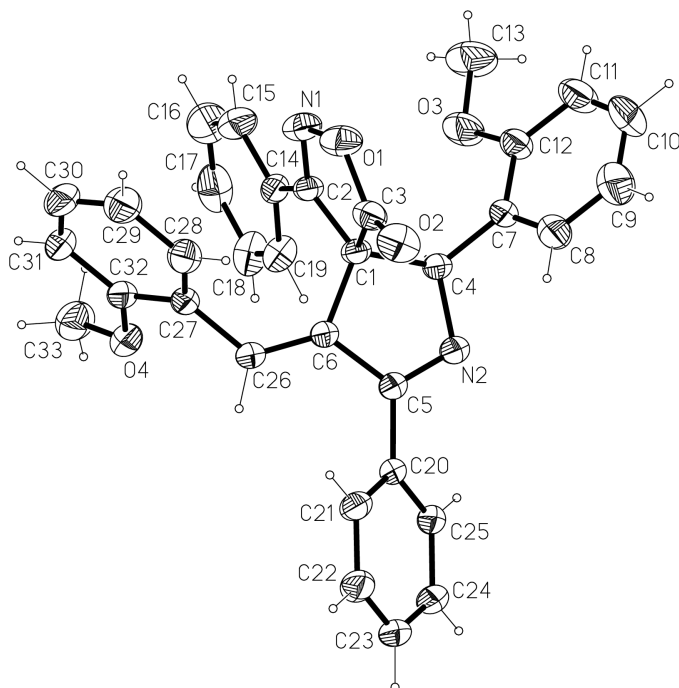
The formation of the title compound, (I), has been interpreted, like the *p*-MeC<sub>6</sub>H<sub>4</sub>- derivative (Bruno *et al.*, 2001a), as the unexpected product of a series of tandem reactions involving Michael- and retro-Michael reactions, C-alkylation, aldol addition and diastereospecific cyclization (Risitano *et al.*, 2001).

The stereochemistry of the C=C double bond and of the tetrahedral C atoms of the product could not easily be established by any spectral technique. The structure in the solid state has been unambiguously determined by X-ray diffraction.



The title compound is very similar to the already reported *p*-Me derivative (Bruno *et al.*, 2001a), obtained by a similar reaction with the same *N*-ylide. The molecular skeleton is a spiro[4.4] system composed of an isoxazolone and a three-substituted pyrroline ring. The two chiral C atoms, the spiro center C1 and the pyrroline C4 atom, have the same configuration (*R,R* in Fig. 1). Due to the crystallographic centre of symmetry, the crystal is the racemic mixture of both enantiomers.

The geometric values of the two similar compounds are comparable within experimental error and are in good agreement with known data (Bertolasi *et al.*, 1994; Brehm *et al.*, 1992). The carbonyl group shows almost the same asymmetry, in agreement with other known carbonyl groups (Allen *et al.*, 1991) and cumarine derivatives (Bruno *et al.*, 2001b).



**Figure 1**  
Perspective view of compound (I) with the atomic numbering scheme. The non-H-atom displacement ellipsoids are at the 30% probability level.

Both compounds have the isoxazolone linked through the C2 atom to a phenyl group.

Although the torsion angles defining the orientation of phenyl with respect to the heterocyclic ring are very different [ $N1-C2-C14-C15 = 0.6$  (3) *versus*  $-32.1$  (5) $^\circ$  in the *p*-tolyl derivative] the C2–C14 bond distances are almost equal. The largest discrepancy between the two similar structures is observed for the C6–C26–C27–C28 torsion angle. Due to steric interactions, the methoxy groups are differently oriented with respect to the phenyl to which they are bonded [ $C13-O3-C12-C7 = 179.4$  (2) $^\circ$ , while  $C33-O4-C32-C27 = -163.8$  (2) $^\circ$ ]. Weak intra- and intermolecular hydrogen-bond interactions determine the crystal packing and the relative orientations of substituents with respect to the single spiro fragment.

## Experimental

The title compound was obtained as a single diastereomer from 4-arylmethyleneisoxazol-5-one and *in situ* prepared *N*-ylide (Risitano *et al.*, 1997) (see reaction Scheme above).

### Crystal data

$C_{33}H_{26}N_2O_4$   
 $M_r = 514.56$   
Triclinic,  $P\bar{1}$   
 $a = 9.127$  (2)  $\text{\AA}$   
 $b = 10.450$  (3)  $\text{\AA}$   
 $c = 14.632$  (3)  $\text{\AA}$   
 $\alpha = 99.172$  (19) $^\circ$   
 $\beta = 101.131$  (17) $^\circ$   
 $\gamma = 101.239$  (18) $^\circ$   
 $V = 1314.6$  (5)  $\text{\AA}^3$

$Z = 2$   
 $D_x = 1.300$   $\text{Mg m}^{-3}$   
Mo  $K\alpha$  radiation  
Cell parameters from 33 reflections  
 $\theta = 6.0$ – $12.9^\circ$   
 $\mu = 0.09$   $\text{mm}^{-1}$   
 $T = 293$  (2) K  
Irregular, colourless  
 $0.17 \times 0.13 \times 0.10$  mm

### Data collection

Siemens P4 diffractometer  
 $\omega$ – $2\theta$  scans  
6449 measured reflections  
6067 independent reflections  
2324 reflections with  $I > 2\sigma(I)$   
 $R_{\text{int}} = 0.032$   
 $\theta_{\text{max}} = 27.6^\circ$

$h = 0 \rightarrow 11$   
 $k = -13 \rightarrow 13$   
 $l = -19 \rightarrow 18$   
3 standard reflections  
every 197 reflections  
intensity decay: none

### Refinement

Refinement on  $F^2$   
 $R[F^2 > 2\sigma(F^2)] = 0.041$   
 $wR(F^2) = 0.097$   
 $S = 0.72$   
6067 reflections  
353 parameters  
H atoms treated by a mixture of independent and constrained refinement

$w = 1/[\sigma^2(F_o^2) + (0.0434P)^2]$   
where  $P = (F_o^2 + 2F_c^2)/3$   
 $(\Delta/\sigma)_{\text{max}} < 0.001$   
 $\Delta\rho_{\text{max}} = 0.14$   $\text{e \AA}^{-3}$   
 $\Delta\rho_{\text{min}} = -0.12$   $\text{e \AA}^{-3}$   
Extinction correction: *SHELXL97*  
Extinction coefficient: 0.0066 (10)

**Table 1**

Selected geometric parameters ( $\text{\AA}$ ,  $^\circ$ ).

O1–C3	1.364 (2)	C1–C2	1.498 (2)
O1–N1	1.440 (2)	C1–C3	1.512 (3)
O2–C3	1.187 (2)	C1–C6	1.520 (2)
O3–C12	1.369 (3)	C1–C4	1.594 (2)
O3–C13	1.417 (2)	C4–C7	1.503 (3)
O4–C32	1.367 (2)	C5–C20	1.473 (2)
O4–C33	1.425 (2)	C5–C6	1.488 (2)
N1–C2	1.287 (2)	C6–C26	1.327 (2)
N2–C5	1.289 (2)	C26–C27	1.480 (2)
N2–C4	1.470 (2)		
C12–O3–C13	118.61 (19)	O2–C3–O1	121.9 (2)
C32–O4–C33	117.50 (16)	O2–C3–C1	129.93 (19)
C2–C1–C3	100.42 (15)	O3–C12–C11	124.8 (2)
C2–C1–C6	120.68 (16)	O3–C12–C7	115.1 (2)
C3–C1–C6	110.80 (16)	C6–C26–C27	125.27 (18)
C2–C1–C4	112.84 (15)	O4–C32–C31	123.5 (2)
C3–C1–C4	110.79 (16)	O4–C32–C27	115.87 (17)
N2–C4–C7–C8	–31.9 (3)	C6–C5–C20–C21	–40.4 (3)
C13–O3–C12–C7	179.44 (19)	C6–C26–C27–C28	–64.1 (3)
N1–C2–C14–C15	0.6 (3)	C33–O4–C32–C27	–163.81 (18)

**Table 2**

Hydrogen-bonding geometry ( $\text{\AA}$ ,  $^\circ$ ).

$D-H\cdots A$	$D-H$	$H\cdots A$	$D\cdots A$	$D-H\cdots A$
C4–H4A $\cdots$ O3	0.98	2.32	2.725 (2)	103
C15–H15A $\cdots$ N1	0.93	2.51	2.810 (3)	99
C30–H30A $\cdots$ O1 <sup>i</sup>	0.93	2.54	3.396 (3)	153

Symmetry code: (i)  $-x, 1-y, -z$ .

Reflection intensities were measured by profile fitting of a 96-step peak scan over  $2\theta$  shells procedure (Diamond, 1969) and then corrected for Lorentz–polarization effects. Standard uncertainties  $\sigma(I)$  were estimated from counting statistics. H atoms were located in idealized positions and allowed to ride on their parent C atoms with isotropic displacement parameters related to the refined values of their corresponding parent atoms.

Data collection: *P3/V* (Siemens, 1989); cell refinement: *P3/V*; data reduction: *SHELXTL-Plus* (Sheldrick, 1990); program(s) used to solve structure: *SIR92* (Altomare *et al.*, 1994); program(s) used to refine structure: *SHELXL97* (Sheldrick, 1997); molecular graphics:

*XPW* (Siemens, 1996); software used to prepare material for publication: *PARST97* (Nardelli, 1995) and *SHELXL97*.

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