

Related literature. (3) was prepared by thermolysis of 2-(methoxybenzyl)-2-phenyl-4-trifluoromethyl-5(2*H*)-oxazolone, (2), which in turn is available from 5-(4-methoxybenzyloxy)-2-phenyl-4-trifluoromethyl-2*H*-oxazolone, (1), by a thermally induced rearrangement (Burger, Schierlinger, Gaa, Geith, Sewald & Müller, 1989). The reaction sequence (1) > (2) > (3) is a unique combination of remarkable reaction steps: the rearrangement (1) > (2) represents a non-concerted 1,5-benzyl-group migration from oxygen to carbon (Burger, Gaa, Geith & Schierlinger, 1989). The transformation (2) > (3) involves a nitrile ylide intermediate which is formed by [3 + 2] cyclo-elimination of carbon dioxide from (2). The

1,3-dipolar species surprisingly acts as a carbene and inserts into the donor substituted aryl moiety to form a cycloheptatriene ring system *via* [2 + 1] cycloaddition and electrocyclic ring opening. Finally, during work-up the 1-pyrroline substructure undergoes an autoxidation process to give compound (3).

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Structure of the 1:1 Adduct of 2-Phenyl-4-(4,6-dimethyl-2-pyrimidinyl)aminomethylene-5(4*H*)-oxazolone and Acetic Acid

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Abstract. $C_{16}H_{14}N_4O_2 \cdot C_2H_4O_2$, $M_r = 354.4$, monoclinic, $P2_1/c$, $a = 12.768$ (2), $b = 7.416$ (3), $c = 19.206$ (3) Å, $\beta = 98.71$ (1)°, $V = 1797.6$ (8) Å³, $Z = 4$, $D_x = 1.309$ g cm⁻³, $\lambda(\text{Mo } K\alpha) = 0.71069$ Å, $\mu = 0.887$ cm⁻¹, $F(000) = 744$, $T = 293$ (2) K, final $R = 0.044$ and $wR = 0.069$ for 1441 observed reflections. The phenyl, pyrimidinyl and oxazolone rings are planar to within 0.010 (5), 0.009 (4) and 0.009 (4) Å, respectively and the whole molecule to within 0.117 (4) Å. The molecule of acetic acid seems to stabilize the planar shape *via* two hydrogen bonds of the type O—H...N [2.756 (5) Å] and O...H—N

[2.891 (5) Å]. The bond lengths and angles agree with expected values.

Experimental. The crystals of the title compound were prepared in an acetic acid solution. A plate-like colourless single crystal with poor scattering properties (0.80 × 0.60 × 0.16 mm) was sealed, together with a drop of mother liquor, into the glass capillary (crystals without the liquid decomposed when exposed to air). Cell dimensions were determined from the angular settings of 25 reflections with $9 < \theta < 12^\circ$ measured on an Enraf-Nonius CAD-4 diffrac-

Table 1. Final fractional coordinates with *e.s.d.*'s in parentheses and equivalent isotropic temperature factors (\AA^2)

U_{eq} is defined as one third of the trace of the orthogonalized U_{ij} tensor.

	x	y	z	U_{eq}
O1	0.8211 (2)	0.0816 (4)	-0.0067 (1)	0.067
C2	0.7796 (3)	0.0547 (5)	0.0549 (2)	0.053
N3	0.6824 (2)	0.1006 (4)	0.0530 (2)	0.051
C4	0.6503 (3)	0.1679 (6)	-0.0145 (2)	0.053
C5	0.7368 (3)	0.1547 (7)	-0.0547 (2)	0.071
O5	0.7480 (3)	0.1922 (6)	-0.1136 (2)	0.108
C6	0.5540 (3)	0.2396 (6)	-0.0376 (2)	0.054
N7	0.4768 (2)	0.2541 (4)	0.0023 (2)	0.049
C8	0.3787 (3)	0.3324 (5)	-0.0207 (2)	0.048
N9	0.3644 (2)	0.4036 (4)	-0.0846 (2)	0.051
C10	0.2695 (3)	0.4782 (6)	-0.1054 (2)	0.054
C13	0.2520 (4)	0.5644 (8)	-0.1767 (3)	0.074
C11	0.1922 (3)	0.4756 (6)	0.0622 (3)	0.066
C12	0.2141 (3)	0.3996 (6)	0.0033 (2)	0.063
C14	0.1369 (4)	0.3864 (11)	0.0535 (3)	0.095
N13	0.3107 (2)	0.3257 (5)	0.0256 (2)	0.059
C31	0.8487 (3)	-0.0251 (5)	0.1146 (2)	0.052
C32	0.8060 (3)	-0.0608 (6)	0.1754 (2)	0.061
C33	0.8659 (4)	-0.1411 (7)	0.2323 (2)	0.070
C34	0.9700 (4)	-0.1895 (7)	0.2289 (3)	0.071
C35	1.0127 (3)	-0.1518 (7)	0.1700 (3)	0.074
C36	0.9535 (3)	-0.0691 (6)	0.1120 (2)	0.062
O11	0.5044 (3)	0.1611 (6)	0.1501 (2)	0.098
O21	0.3606 (2)	0.2965 (6)	0.1699 (2)	0.101
C21	0.4524 (4)	0.2241 (7)	0.1909 (2)	0.067
C22	0.4855 (6)	0.2292 (9)	0.2683 (3)	0.092

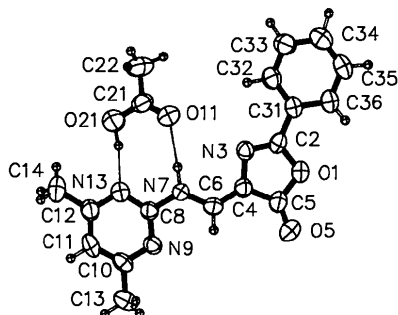


Fig. 1. ORTEP (Johnson, 1965) view of the molecule with the atom-numbering scheme. Thermal ellipsoids for the non-H atoms are drawn at 50% probability level.

tometer. Space group $P2_1/c$ was determined from the systematic absences. Intensity data were collected in the range $1 < 2\theta < 54^\circ$ with variable speed $\omega-2\theta$ scans using graphite-monochromated Mo $K\alpha$ radiation: max. scan time 30 s, scan width $(0.8 + 0.3\tan\theta)^\circ$ and aperture $(2.4 + 0.9\tan\theta)$ mm. The background was measured for 1/4 of the scan time at each scan limit. Three standard reflections were monitored at intervals of 500 reflections. The crystal orientation was checked every 2 h. Data were corrected for intensity variation (-2.2%) and Lp effects, but absorption was ignored. 3910 unique reflections were collected of which 1441 were considered observed [$I > 2\sigma(I)$, $\sigma(I)$ based on counting statistics] and used

Table 2. Bond distances (\AA) and angles ($^\circ$) with *e.s.d.*'s in parentheses

O1—C2	1.383 (5)	C10—C11	1.383 (7)
O1—C5	1.414 (5)	C11—C12	1.368 (7)
C2—N3	1.282 (5)	C12—C14	1.483 (7)
C2—C31	1.461 (6)	C12—N13	1.358 (5)
N3—C4	1.392 (6)	C31—C32	1.387 (6)
C4—C5	1.443 (6)	C31—C36	1.385 (6)
C4—C6	1.351 (6)	C32—C33	1.373 (6)
C5—O5	1.194 (6)	C33—C34	1.388 (8)
C6—N7	1.341 (5)	C34—C35	1.357 (8)
N7—C8	1.391 (5)	C35—C36	1.390 (7)
C8—N9	1.324 (6)	O11—C21	1.196 (6)
C8—N13	1.333 (6)	O21—C22	1.297 (7)
N9—C10	1.337 (5)	C21—C22	1.484 (7)
C10—C13	1.497 (7)		
C2—O1—C5	105.1 (3)	N9—C10—C11	120.8 (4)
O1—C2—N3	115.6 (4)	C13—C10—C11	122.3 (4)
O1—C2—C31	117.8 (3)	C10—C11—C12	119.5 (4)
N3—C2—C31	126.6 (3)	C11—C12—C14	124.1 (4)
C2—N3—C4	105.6 (4)	C11—C12—N13	120.4 (4)
N3—C4—C5	109.4 (4)	C14—C12—N13	115.5 (4)
N3—C4—C6	124.9 (3)	C8—N13—C12	115.2 (4)
C5—C4—C6	125.7 (3)	C2—C31—C32	118.1 (4)
O1—C5—C4	104.3 (3)	C2—C31—C36	122.6 (3)
O1—C5—O5	121.2 (4)	C32—C31—C36	119.3 (4)
C4—C5—O5	134.5 (3)	C31—C32—C33	120.7 (4)
C4—C6—N7	123.7 (4)	C32—C33—C34	119.8 (4)
C6—N7—C8	123.9 (3)	C33—C34—C35	119.7 (5)
N7—C8—N9	116.8 (3)	C34—C35—C36	121.3 (4)
N7—C8—N13	114.8 (4)	C31—C36—C35	119.1 (4)
N9—C8—N13	128.4 (3)	O11—C21—O21	121.6 (4)
C8—N9—C10	115.7 (3)	O11—C21—C22	124.4 (5)
N9—C10—C13	116.9 (4)	O21—C21—C22	114.1 (5)
O21—H21	0.97 (5)	N7—H7	0.95 (5)
H21...N13	1.79 (5)	H7...O11	1.94 (5)
O21...N13	2.756 (5)	N7...O11	2.891 (5)
O21—H21...N13	171.3 (6)	N7—H7...O11	177.8 (1)
C12—N13...O21	114.1 (2)	C8—N7...O11	112.4 (2)
C8—N13...O21	126.8 (2)	C6—N7...O11	123.4 (2)
C21—O21...N13	113.9 (3)	C21—O11...N7	123.6 (3)

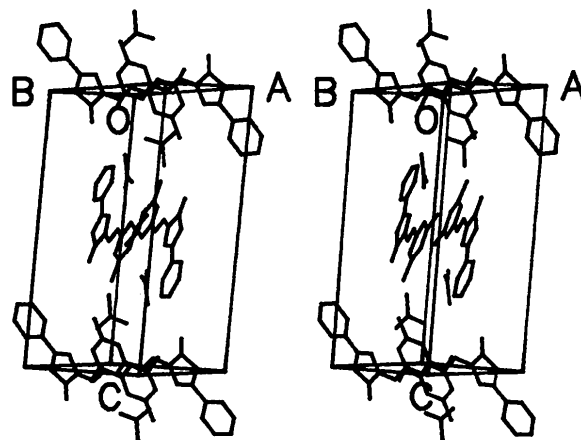


Fig. 2. Stereoview of the molecular packing.

in the refinement; range of hkl $h -13/13$, $k 0/8$, $l 0/22$. The structure was solved with *MULTAN80* (Main, Fiske, Hull, Lessinger, Germain, Declercq & Woolfson, 1980). The initial structural model was

refined by full-matrix least squares minimizing the function $\sum w(|F_o| - |F_c|)^2$ with *SHELX76* (Sheldrick, 1976). Final *R* and *wR* values were 0.044 and 0.069, respectively with $w = 0.260/[\sigma^2(F_o) + 0.0094F_o^2]$ and 293 variables. The H-atom positions were found in $\Delta\rho$ map and included in the refinement with a constraint imposed on the C—H bonds. The common isotropic temperature factor *U* for methyl H atoms was 0.151 (9) Å² and for other H atoms was 0.071 (4) Å². The final $\Delta\rho$ showed residual electron density within +0.12 and -0.08 e Å⁻³. Max. value of Δ/σ at convergence was 0.62 (*z*, H142). Scattering factors for C, N and O were from *International Tables for X-ray Crystallography* (1974, Vol. IV), for H from Stewart, Davidson & Simpson (1965). All calculations performed on the VAX 8550 under VMS 5.2 at University Computer Centre, Ljubljana. Additionally, the *GX* (Mallinson & Muir, 1985) package was used for data processing and final interpretation of molecular geometry. The final atomic parameters are given in Table 1.* Bond lengths and angles are given in Table 2. Views of the molecule with the atomic numbering and of the molecular packing are presented in Figs. 1 and 2.

* Lists of structure factors, anisotropic thermal parameters, H-atom coordinates, selected torsion angles and the results of calculated mean-planes angles have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 53890 (13 pp.). Copies may be obtained through The Technical Editor, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Related literature. Recently Stanovnik, Svete & Tišler (1987), Stanovnik, Svete, Tišler, Žorž, Hvala & Simonič, (1988) and Svete, Stanovnik, Tišler, Golič & Leban (1989) have described the syntheses of β -heteroaryl amino- α,β -dehydro- α -amino acids and their derivatives from *N*-heteroaryl formamidines and 2-phenyl-5-oxo-1,3-oxazole in which 4-heteroarylaminomethylene-2-phenyl-5(4*H*)-oxazolones are formed as novel intermediates.

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Structure of Methyl (4*aS*,5*R*,7*aR*,11*aR*,12*aS*)-2-Benzyl-2,3,4,4*a*,5,6,7,7*a*,8,10,11*a*,12-dodecahydro-7,7-(ethylenedioxy)-1,9-dioxooxazolo[3',4':1,2]pyrrolo[5,4-*i*]isoquinoline-5-carboxylate Toluene (2/1)

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Abstract. 2C₂₄H₂₈N₂O₇·C₇H₈, *M_r* = 502.57, triclinic, *P*1, *a* = 11.076 (1), *b* = 11.314 (1), *c* = 12.155 (1) Å, α = 104.41 (1), β = 102.02 (1), γ = 89.37 (1)°, *V* = 1441.6 (3) Å³, *Z* = 1, *D_x* = 1.16 g cm⁻³, $\lambda(\text{Cu K}\alpha)$ = 1.5418 Å, μ = 6.55 cm⁻¹, *F*(000) = 534, room temperature, final *R* = 0.106 for 5273 observed reflections. The asymmetric unit contains two independent molecules, *A* and *B*, of the title compound C₂₄H₂₈-

N₂O₇ plus one toluene molecule. Molecules *A* and *B* show similar conformations except for different orientations of the carboxylate group [O(6)—C(16)—O(7)—C(17)]: torsion angles C(3)—C(4)—C(16)—O(6) are -62 (2)° and +111 (1)° for *A* and *B* respectively. Excluding the carboxylate group least-squares matching of the positions of corresponding non-H atoms of *A* and *B* yields a r.m.s. deviation of 0.20 Å.