$S = 1.037$
5619 reflections
253 parameters
H-atom parameters constrained

$\Delta\rho_{\max} = 0.20$ e Å^{-3}
$\Delta\rho_{\min} = -0.21$ e Å^{-3}
Extinction correction: none
Scattering factors from *International Tables for Crystallography* (Vol. C)

Table 1. *Selected geometric parameters (Å, °)*

S1—O3	1.4250 (14)	N2′—C3′	1.262 (3)
S1—O2	1.439 (2)	N3—C2	1.482 (2)
S1—N3	1.628 (2)	N3—C4	1.497 (2)
S1—C11	1.759 (2)	C2—C5′	1.518 (3)
O1—C2	1.402 (2)	C3′—C4′	1.478 (3)
O1—C5	1.427 (2)	C4—C5	1.538 (3)
O1′—N2′	1.428 (2)	C4′—C5′	1.514 (3)
O1′—C5′	1.444 (2)		
O3—S1—O2	120.95 (9)	O1—C2—C5′	110.08 (15)
O3—S1—N3	106.73 (9)	N3—C2—C5′	113.20 (15)
O2—S1—N3	105.49 (9)	N2′—C3′—C4′	115.7 (2)
O3—S1—C11	108.08 (9)	N2′—C3′—C31	120.4 (2)
O2—S1—C11	106.84 (10)	C4′—C3′—C31	123.9 (2)
N3—S1—C11	108.21 (9)	N3—C4—C41	110.2 (2)
C2—O1—C5	105.87 (13)	N3—C4—C5	100.45 (14)
N2′—O1′—C5′	109.16 (14)	C3′—C4′—C5′	100.8 (2)
C3′—N2′—O1′	108.0 (2)	O1—C5—C4	103.08 (14)
C2—N3—C4	108.23 (13)	O1′—C5′—C4′	104.9 (2)
C2—N3—S1	119.65 (11)	O1′—C5′—C2	107.54 (15)
C4—N3—S1	119.92 (12)	C4′—C5′—C2	114.9 (2)
O1—C2—N3	104.14 (13)		

C5′—O1′—N2′—C3′	8.2 (2)
C5—O1—C2—N3	−37.0 (2)
C5—O1—C2—C5′	−158.67 (15)
C4—N3—C2—O1	15.1 (2)
C4—N3—C2—C5′	134.7 (2)
O1′—N2′—C3′—C4′	−0.3 (2)
C2—N3—C4—C41	−112.3 (2)
C2—N3—C4—C5	10.4 (2)
N2′—C3′—C4′—C5′	−7.1 (2)
C2—O1—C5—C4	44.2 (2)
N3—C4—C5—O1	−32.0 (2)
C41—C4—C5—O1	86.6 (2)
N2′—O1′—C5′—C4′	−12.2 (2)
N2′—O1′—C5′—C2	110.5 (2)
C3′—C4′—C5′—O1′	11.1 (2)
C3′—C4′—C5′—C2	−106.8 (2)
O1—C2—C5′—O1′	−71.5 (2)
N3—C2—C5′—O1′	172.37 (13)
O1—C2—C5′—C4′	44.8 (2)
N3—C2—C5′—C4′	−71.3 (2)

The H atoms were introduced at calculated positions (C—H distances 0.93–0.98 Å, depending on atom type) and treated as riding atoms, with displacement parameters 1.2 times the U_{eq} value of the parent atom.

Data collection: *XSCANS* (Siemens, 1996). Cell refinement: *XSCANS*. Data reduction: *SHELXTL* (Sheldrick, 1995). Program(s) used to solve structure: *SHELXS86* (Sheldrick, 1990). Program(s) used to refine structure: *SHELXL93* (Sheldrick, 1993). Molecular graphics: *SHELXTL*. Software used to prepare material for publication: *SHELXTL*.

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2,3,4,5,6,7-Hexahydro-9,10-dimethoxy-1,2-benzothiazonin-3-one 1,1-dioxide

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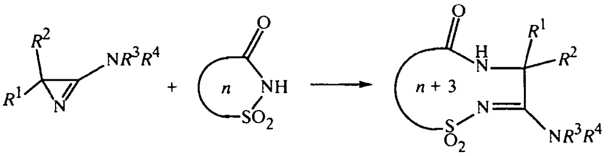
Abstract

The amide group in the nine-membered ring of the title compound, $C_{13}H_{17}NO_5S$, has the *trans* conformation. The molecules are linked into infinite one-dimensional chains by bifurcated intermolecular N—H···O hydrogen bonds involving the amide O atom and one of the sulfonyl O atoms of the same neighbouring molecule. The compound was prepared by the Friedel–Crafts acylation of 1,2-dimethoxybenzene and glutaric acid anhydride, followed by reduction of the aryl ketone, esterification, chlorosulfonation, treatment with ammonia, saponification and cyclization of the corresponding 5-(2-sulfamoylphenyl)butanoic acid.

Comment

For several years, we have been studying ring enlargement reactions of NH acidic heterocycles using 3-amino-2*H*-azirines, (I), as reagents (Heimgartner, 1991). For a successful reaction, the pK_a value of the heterocycle has to be below 8 as the first reaction step is the protonation of (I). This requires the heterocyclic NH group to be flanked by electron-withdrawing groups. One class of compounds which meets this requirement is that of the cyclic sulfonamides (sultams) of type (II). The scope and limitations of this ring enlargement have been established by conducting a series of reactions of (I) with four- to nine-membered oxosulfonamides, (II), in which the corresponding seven- to twelve-membered heterocycles, (III), are obtained as the products (Heimgartner, 1991; Orahovats *et al.*, 1992, 1996; Mihova *et al.*, 1996, 1998; Linden *et al.*, 1998).

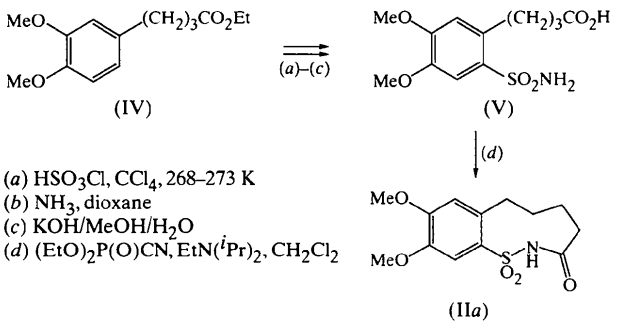


(I) (II) (III)

Some five-membered cyclic oxosulfonamides of type (II) are well known. Saccharin is one example (Fahlberg & Remsen, 1879; Hettler, 1973) and the crystal structure of saccharin was published by independent workers 30 years ago (Bart, 1968; Okaya, 1969). The molecules are linked into centrosymmetric dimers by intermolecular N—H···O hydrogen bonds between the amide groups of adjacent molecules. Similar structures have been found for a four-membered (Mihova *et al.*, 1996) and an eight-membered analogue (Mihova *et al.*, 1998).

Whereas in the case of the four- to seven-membered oxosulfonamides, the ring is built up by treatment of the corresponding sulfonamidocarboxylic acids of type (V) in benzene with PCl_5, similar cyclizations to the analogous eight- and nine-membered rings failed (Mihova *et al.*, 1998). A successful ring closure of (V) to give the nine-membered ring in the form of the title compound, (II*a*), in 40.1% yield was performed using diethylphosphoryl cyanide (DEPC) in the presence of ethyldiisopropylamine (*cf.* Yamada *et al.*, 1973). The analogous eight-membered ring was obtained in 62.4% yield (Mihova *et al.*, 1998).

The low-temperature structure determination of (II*a*) shows that the bond lengths and angles have values normally observed in this class of compounds. In contrast to the eight-membered analogue, in which the amide bond has the *cis* conformation, the conformation about the amide bond (S1—N2—C3—C4) of the nine-membered ring in (II*a*) lies closer to the *trans* conformation (Fig. 1). This conformational arrangement means that it is not possible for hydrogen-bonded dimers to exist in the crystal. Instead, the amide NH group forms bifurcated intermolecular hydrogen bonds, with the amide O atom and one of the sulfonyl O atoms of the same neighbouring molecule acting as the acceptor atoms (Table 2). These double hydrogen-bonding bridges link the molecules into infinite one-dimensional chains which run parallel to the [100] direction and each interaction has a unitary graph-set motif of *C*(4) (Bernstein *et al.*, 1995). The local ring formed by the bifurcated system has a binary graph-set motif of $R_1^2(6)$.



(*a*) HSO_3Cl, CCl_4, 268–273 K
(*b*) NH_3, dioxane
(*c*) KOH/MeOH/H_2O
(*d*) $(EtO)_2P(O)CN$, $EtN(^iPr)_2$, CH_2Cl_2

The nine-membered ring is quite puckered (Table 1) and all ring atoms from N2 to C6, inclusive, lie on the same side of the plane of the benzo group. Thus, the overall molecular geometry is sharply angular, with the bend occurring along the S1···C7 axis. Excluding C8 and C13, the mean plane of the ring atoms of the nine-membered ring (r.m.s. deviation 0.435 Å) makes an angle of 59.69 (5)° with the mean plane of the benzo moiety, the calculation of which includes the coplanar methoxy groups, as well as S1 and C7. The r.m.s. deviation of the constituent atoms from this latter plane is 0.025 Å, with a maximum deviation of 0.039 (2) Å for C14.

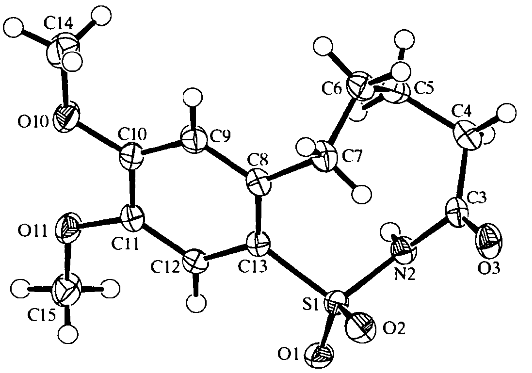


Fig. 1. View of the molecule of (II*a*) showing the atom-labelling scheme. Displacement ellipsoids are drawn at the 50% probability level and H atoms are represented by circles of arbitrary size.

Experimental

The title compound, (II*a*), was obtained in 40.1% yield by stirring a diluted (0.5 mmol/50 ml CH_2Cl_2) 1:1 mixture of 5-(4,5-dimethoxy-2-sulfamoylphenyl)butanoic acid, (V), and diethylphosphoryl cyanide in the presence of excess ethyl(diisopropyl)amine in dichloromethane at room temperature for 20 h (Mihova *et al.*, 1998). Chromatographic separation on silica gel and recrystallization from dichloromethane–hexane gave colourless tablets (m.p. 464.5–466.5 K).

Crystal data

$C_{13}H_{17}NO_5S$
$M_r = 299.34$
Orthorhombic
Pbca
$a = 9.165\ (3)$ Å
$b = 36.319\ (3)$ Å
$c = 8.353\ (4)$ Å
$V = 2780.3\ (15)$ Å^3
$Z = 8$
$D_x = 1.430$ Mg m^{-3}
D_m not measured

Mo $K\alpha$ radiation
$\lambda = 0.71069$ Å
Cell parameters from 24 reflections
$\theta = 18–20°$
$\mu = 0.251$ mm^{-1}
$T = 213\ (1)$ K
Tablet
0.45 × 0.38 × 0.13 mm
Colourless

Data collection

Rigaku AFC-5*R* diffractometer
ω scans
Absorption correction: none
4242 measured reflections
3174 independent reflections
2338 reflections with $I > 2\sigma(I)$

$R_{\rm int} = 0.020$
$\theta_{\rm max} = 27.5°$
$h = 0 \rightarrow 11$
$k = -1 \rightarrow 47$
$l = -1 \rightarrow 10$
3 standard reflections every 150 reflections intensity decay: none

Refinement

Refinement on F^2
$R[F^2 > 2\sigma(F^2)] = 0.042$
$wR(F^2) = 0.115$
$S = 1.040$
3174 reflections
187 parameters
H atoms: see below
$w = 1/[\sigma^2(F_o^2) + (0.048P)^2 + 0.866P]$ where $P = (F_o^2 + 2F_c^2)/3$

$(\Delta/\sigma)_{\rm max} = 0.001$
$\Delta\rho_{\rm max} = 0.43$ e Å^{-3}
$\Delta\rho_{\rm min} = -0.37$ e Å^{-3}
Extinction correction: none
Scattering factors from *International Tables for Crystallography* (Vol. C)

Table 1. *Selected geometric parameters (Å, °)*

S1—N2	1.6500 (17)	O3—C3	1.212 (2)
S1—C13	1.775 (2)	N2—C3	1.415 (3)
C13—S1—N2—C3	91.83 (18)	C5—C6—C7—C8	−60.2 (2)
S1—N2—C3—O3	44.7 (3)	C6—C7—C8—C13	108.5 (2)
S1—N2—C3—C4	−133.63 (17)	C9—C8—C13—S1	178.17 (15)
N2—C3—C4—C5	61.3 (2)	N2—S1—C13—C8	−60.29 (19)
C3—C4—C5—C6	59.8 (2)	H2—N2—C3—O3	−167 (2)
C4—C5—C6—C7	−72.3 (2)		

Table 2. *Hydrogen-bonding geometry (Å, °)*

D—H$\cdots A$	D—H	H$\cdots A$	$D\cdots A$	D—H$\cdots A$
N2—H2$\cdots$O2$^{\rm i}$	0.84 (2)	2.09 (2)	2.841 (2)	150 (2)
N2—H2$\cdots$O3$^{\rm i}$	0.84 (2)	2.47 (2)	3.114 (2)	134 (2)

Symmetry code: (i) $\frac{1}{2} + x, y, \frac{1}{2} - z$.

The position of the amide H atom was refined freely together with an isotropic displacement parameter. Methyl H atoms were located from a difference Fourier synthesis and were constrained to an ideal geometry with $U_{\rm iso}({\rm H}) = 1.5U_{\rm eq}({\rm C})$, but were allowed to rotate freely about the C—O bonds. The positions of all other H atoms were geometrically idealized and were refined using a riding model with $U_{\rm iso}({\rm H}) = 1.2U_{\rm eq}({\rm C})$

Data collection: *MSC/AFC Diffractometer Control Software* (Molecular Structure Corporation, 1991). Cell refinement: *MSC/AFC Diffractometer Control Software*. Data reduction: *TEXSAN* (Molecular Structure Corporation, 1997). Program(s) used to solve structure: direct methods *SHELXS*97 (Sheldrick, 1997*a*). Program(s) used to refine structure: *SHELXL*97 (Sheldrick, 1997*b*). Molecular graphics: *ORTEP*II (Johnson, 1976). Software used to prepare material for publication: *SHELXL*97.

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Supplementary data for this paper are available from the IUCr electronic archives (Reference: BM1336). Services for accessing these data are described at the back of the journal.

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