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Formation and Molecular Structure of the 2:1-Adducts of 1-Alkoxy-2-benzopyrylium-4-olates with Carbon Disulfide

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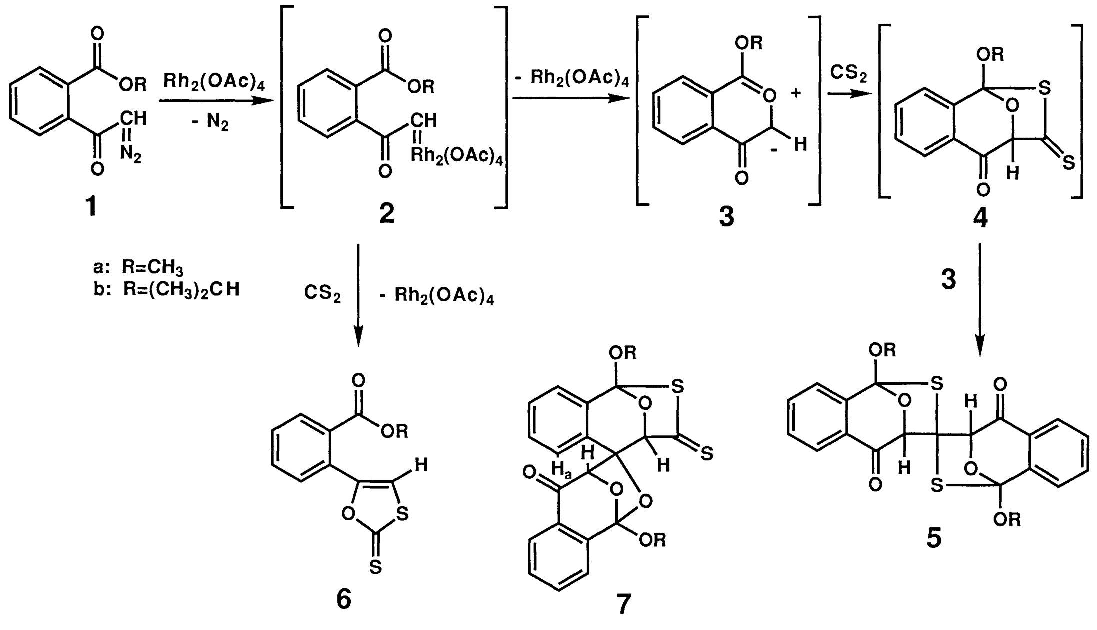
The $Rh_2(OAc)_4$-catalyzed decomposition of alkyl *o*-(diazoacetyl)benzoates in the presence of carbon disulfide gave two 2:1-adducts of the intermediate 1-alkoxy-2-benzopyrylium-4-olates with carbon disulfide together with a small amount of 5-(*o*-alkoxycarbonylphenyl)-1,3-oxathiole-2-thiones. The intramolecular carbene-carbonyl reaction of the carbenoid generated by the decomposition of the diazo ketones was clarified to be much faster than the intermolecular attack on carbon disulfide. The molecular structure of the major 2:1-adduct of 1-methoxy-2-benzopyrylium-4-olate with carbon disulfide was determined by means of X-ray crystallography. The crystals of the adduct are triclinic with space group $P\bar{1}$, and with the unit-cell dimensions of a=12.213(1), b=11.846(1), c=7.0403(2) Å, α=93.395(6), β=102.147(5), γ=109.536(7)°, and Z=2. The final R value is 0.044 for 2597 observed reflections. The structure was confirmed to be (*S-endo*, *S′-endo*).

Recently, formation of carbonyl ylides by carbene–carbonyl reaction,[1)] and their 1,3-dipolar cycloaddition with various types of dipolarophiles have been studied extensively from the synthetic interests.[2)] The authors reported previously the 1,3-dipolar cycloaddition of 1-methoxy-2-benzopyrylium-4-olate (**3a**), which was formed by the intramolecular carbene–carbonyl reaction of the carbenoid **2a**, with ethylenic,[1b,3)] acetylenic,[1b,4)] and carbonyl compounds.[5)] In the continuation of these studies we report here the reaction of **3** with carbon disulfide as an example of dipolarophiles having a C–S double bond.

Results and Discussion

The Catalytic Decomposition of Alkyl *o*-(Diazoacetyl)benzoates (1) in the Presence of Carbon Disulfide. The medium pressure column chromatography of the $Rh_2(OAc)_4$-catalyzed reaction mixture of methyl *o*-(diazoacetyl)benzoate (**1a**) with carbon disulfide at 46 °C gave two 2:1-adducts **5a** of the corresponding carbonyl ylide intermediate **3a** with carbon disulfide along with 5-(*o*-methoxycarbonylphenyl)-1,3-oxathiole-2-thione (**6a**). The formation of **6a** was described in the previous papers.[6,7)]

Elemental analysis of the 2:1-adducts **5a** indicates that they contain two molecules of **3a** and a molecule of carbon disulfide. The NMR spectra of the major 2:1-adduct (**5a-A**; 64.5% yield) showed that two moieties of **3a** are equivalent each other in **5a-A**. For example, the ^{1}H NMR spectrum of **5a-A** has only one signal of methine protons at δ=5.01 and that of methoxyl-protons at δ=3.95. Similarly, the ^{13}C NMR spectrum of **5a-A** also exhibited only one signal for each pairs of methoxyl-carbons (δ=53.41), methine-



Scheme 1.

Table 1. NMR Data of 2:1-Adducts

	Major 2:1-adduct **5a-A**	Minor 2:1-adduct **5a-B**		Major 2:1-adduct **5b-A**	Minor 2:1-adduct **5b-B**
^{1}H NMR ($CDCl_3$)					
OCH_3 (s)	3.95 (6H)	3.74 (3H)	CH_3 (d)	1.43 (6H, J=6.2 Hz)	1.26 (3H, J=6.2 Hz)
		3.87 (3H)			1.39 (3H, J=6.2 Hz)
CH (s)	5.01 (2H)	5.08 (1H)		1.44 (6H, J=6.2 Hz)	1.41 (3H, J=6.2 Hz)
		5.29 (1H)			1.48 (3H, J=6.2 Hz)
arom-H (m)	7.42—7.62 (6H)	7.41—7.63 (6H)	CH (s)	4.98 (2H)	5.04 (1H)
	8.03—8.05 (2H)	7.96—8.03 (2H)			5.30 (1H)
			CH (sept)	5.03 (2H, J=6.2 Hz)	4.51 (1H, J=6.2 Hz)
					4.76 (1H, J=6.2 Hz)
			arom-H (m)	7.40—8.04 (8H)	7.38—8.01 (8H)
^{13}C NMR ($CDCl_3$)					
OCH_3 (q)	63.41	52.52	CH_3 (q)	23.26	23.06
		52.69		23.88	23.31
spiro-C (s)	70.91	75.78			23.71
CH	89.86[a)]	83.62[b)]			23.94
		94.35[c)]	spiro-C (s)	71.00	75.89
S-C-O (s)	117.40	117.06	OCH (d)	71.31	70.77
		117.50			71.04
arom-CH (d)	120.76	120.47	CH	89.58[d)]	83.07[e)]
		120.63			94.17[f)]
arom-C (s)	127.23	127.00	S-C-O (s)	116.97	116.76
		127.60			117.07
arom-CH (d)	127.45	127.71	arom-CH (d)	121.08	120.78
		127.74			120.90
arom-CH (d)	128.85	128.84	arom-C (s)	127.10	126.63
		129.06			127.27
arom-CH (d)	134.77	134.57	arom-CH (d)	127.36	127.55
		134.70			127.60
arom-C (s)	147.77	147.97	arom-CH (d)	128.35	128.62
		148.35			128.86
C=O (s)	188.89	189.97	arom-CH (d)	134.72	134.58
		190.38			134.72
			arom-C (s)	148.34	148.52
					148.79
			C=O (s)	188.97	189.96
					190.26

a) dd, $^{3}J_{CH}$=3.4 Hz. b) d, $^{3}J_{CH}$=0 Hz. c) dd, $^{3}J_{CH}$=3.1 Hz. d) dd, $^{3}J_{CH}$=3.4 Hz. e) d, $^{3}J_{CH}$=0 Hz. f) dd, $^{3}J_{CH}$=3.1 Hz.

carbons (δ=89.86), carbonyl-carbons (δ=188.89), and other carbons of two benzopyrylium-olate moieties as are shown in Table 1. On the other hand, in the minor 2:1-adduct (**5a-B**; 10.8% yield) two **3a** moieties involved are not equivalent each other, and the corresponding protons and carbons of two **3a** moieties show their signals at different positions in its ^{1}H and ^{13}C NMR spectra (Table 1).

Quite similar results were also obtained in the $Rh_2(OAc)_4$-catalyzed decomposition of isopropyl *o*-(diazoacetyl)benzoate (**1b**) in the presence of carbon disulfide.

In these reactions of **1a** and **1b**, no 1:1-adduct **4** between **3** and carbon disulfide was observed in spite of the detailed inspection of the reaction mixture by column chromatography. This indicates that the second cycloaddition of **3** with the 1:1-adducts **4** is much faster than the first attack of **3** on carbon disulfide.

There are two sites of the second attack of **3** in a molecule of the 1:1-adduct **4**. One is thiocarbonyl group to give **5**, and the other is carbonyl group to give **7**. From our previous studies,[5c,8)] **7** should show a signal of an aromatic proton H_a at high field in its ^{1}H NMR spectrum due to the ring current of the neighboring aromatic ring system. The fact that our major and minor 2:1-adducts show no such a high field signal indicates that the second attack of **3** to give these 2:1-adducts did not occur on the carbonyl group but occurred on the thiocarbonyl group of **4**. Absence of a signal of thiocarbonyl carbon in the ^{13}C NMR spectra of these adducts in the region of δ=202—203[7)] also supports the reaction site described above. The regiochemistry of the first and the second cycloadditions of **3** is consistent with the presence of singlet signals of nonaromatic quaternary carbons having orthothioformate structure at near δ=117 in the ^{13}C NMR spectra of **5**.[5)]

These 1,3-dipolar cycloadditions of carbonyl ylides have been found to proceed through the interaction

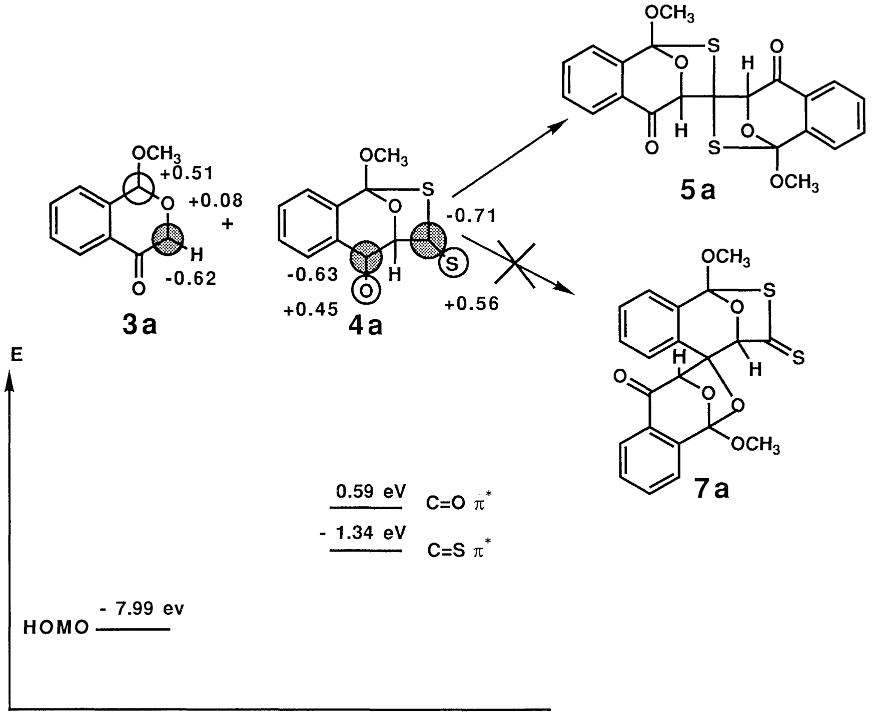
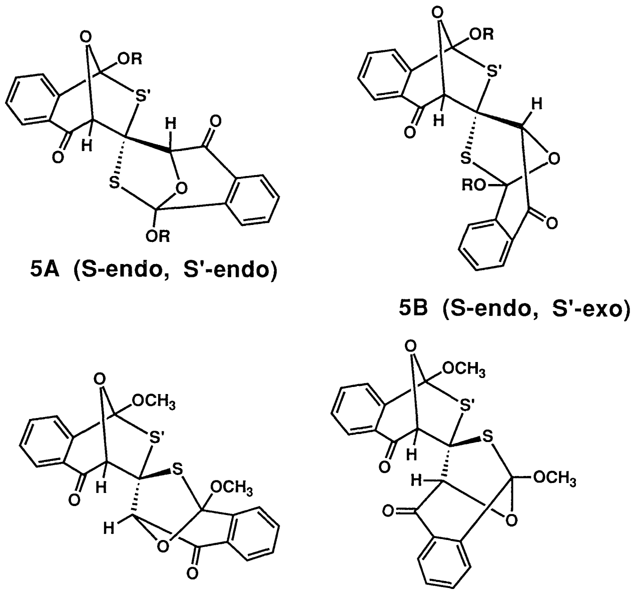


Fig. 1. The energy levels and atomic orbital coefficients of frontier orbitals of **3a** and **4a**.

controlled by carbonyl ylide-HOMO and dipolarophile-LUMO.[5b,9] In order to presume the reaction site of the second attack of **3a** toward 1 : 1-adduct **4a**, energy levels of the C=O π* and C=S π* of **4a** were calculated together with HOMO level of **3a**. Results of MNDOC calculation[10] shown in Fig. 1 revealed that the energy level of C=S π* (−1.34 eV) is lower than that of C=O π* (0.59 eV). This indicates that the reaction site is not the C=O group but the C=S group affording the 2:1-adducts **5** as is described above. The atomic orbital coefficients of the related frontier orbitals of **3a** and **4a** also explain the regiochemistry of the cycloaddition.[5b]

There are four possibilities in the second attack of **3** on C–S double bond of the 1:1-adduct **4** in a regiochemistry similar to that of the first cycloaddition to afford four stereoisomers, **5-A** (S-*endo*, S′-*endo*),[11] **5-B** (S-*endo*, S′-*exo*), **5-C** (S-*exo*, S′-*endo*), and **5-D** (S-*exo*, S′-*exo*) as shown in Fig. 2. In these four isomers, **5-B** and **5-C** are enantiomers each other, and the two benzopyrylium-4-olate moieties of **5-B** (=**5-C**) are not equivalent. Therefore, the minor 2:1-adduct can be assigned to the structure **5-B**. On the other hand, both **5-A** and **5-D** have a symmetric structure having a C_2-axis through spiro-carbon bisecting both S–C–S and CO–C–CO planes. Because the two benzopyrylium-4-olate moieties are equivalent in **5-A** and **5-D**, the structural assignment for the major 2:1-adducts to **5-A** or **5-D** was not successful on the basis of the NMR data. Single crystal X-ray analysis of the major 2:1-adduct **5a-A** was therefore studied.


5A (S-endo, S'-endo)

5B (S-endo, S'-exo)

5C (S-exo, S'-endo)

5D (S-exo, S'-exo)

Fig. 2. Possible four structures of the 2: 1-adduct **5**.

X-Ray Crystallography of the 2:1-Adduct 5a-A. The crystal data of **5a-A** are listed in Table 2 along with the experimental details. The ORTEP[12] drawing (see Fig. 3) shows that the major 2:1-adduct **5a-A** has the symmetric structure **5-A** (S-*endo*, S′-*endo*).

Inspection of the CPK model of **5-A** and **5-D** indicates that carbonyl groups of two benzopyrylium-

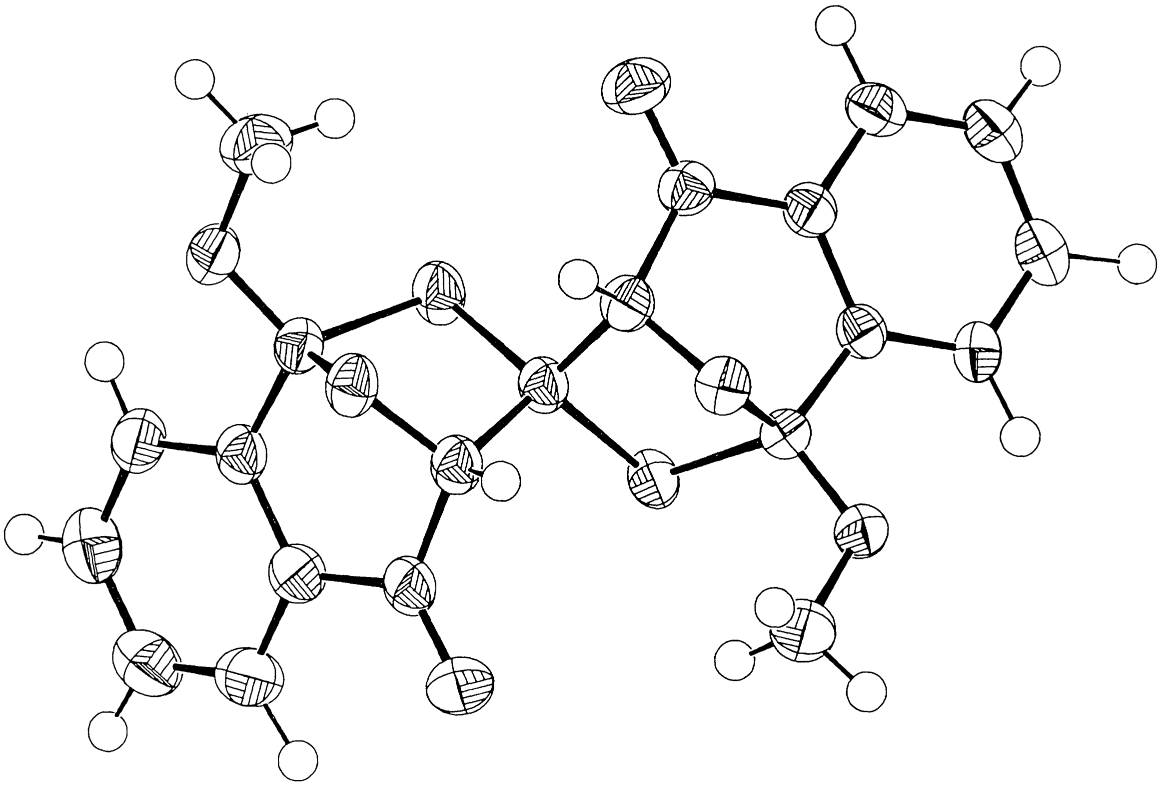


Fig. 3. ORTEP drawing of the 2:1-adduct **5a-A**.

Table 2. Crystal Data and Experimental Details of **5a-A**

Molecular formula	$C_{21}H_{16}O_6S_2$
Molecular weight	428.49
Crystal system	Triclinic
Space group	$P\bar{1}$
a	12.213(1) Å
b	11.846(1) Å
c	7.0403(2) Å
α	93.395(6)°
β	102.147(5)°
γ	109.536(7)°
V	929.1(1) $Å^3$
Z	2
D_m	1.550(2) Mg m^{-3}
D_x	1.532 Mg m^{-3}
μ (Cu $K\alpha$)	28.551 cm^{-1}
Scan method	ω-2θ
Scan speed in ω	4° min^{-1}
Scan width	$(1.1+0.15 \tan\theta)°$
Back ground	2×4 s
Number of reflections	2753
	2597 ($\lvert F_o \rvert \geqq 2\sigma(F_o)$)
R	0.044
Crystal size	0.3×0.2×0.2 mm
$2\theta_{max}$	120°

olate moieties of **5-D** come in the region of van der Waals radii of these groups, and that the approach to give **5-D** is hindered by the steric repulsion. On the other hand, isomers **5-A** and **5-B** (=**5-C**) are free from this sort of steric repulsion. The model of **5-B** is a little crowded than that of **5-A**. These facts will explain the product ratio of the reactions described above.

Experimental

Melting points were measured with a Yanagimoto Melting Point Apparatus and are not corrected. IR spectra were recorded on a Perkin-Elmer Model 983. ^1H NMR and ^{13}C NMR spectra were measured on a Varian EM 390 and a Brucker AM 360 spectrometers in a $CDCl_3$ solution using TMS as an internal standard.

Materials. Methyl *o*-(diazoacetyl)benzoate (**1a**) and isopropyl *o*-(diazoacetyl)benzoate (**1b**) were prepared by the procedure described in the previous paper.[5b)] Benzene and carbon disulfide were purified by distillation over CaH_2 and P_2O_5, respectively.

General Procedure of the $Rh_2(OAc)_4$-Catalyzed Decomposition of 1 in the Presence of CS_2. A benzene solution (10 ml) of **1** (3.00 mmol) was added dropwise into a solution of $Rh_2(OAc)_4$ (1.6 mg, 3.62×10^{-3} mmol) in carbon disulfide (20 ml) at 46 °C under stirring in a N_2 atmosphere. After being refluxed for about 1 h, the reaction mixture was concentrated under reduced pressure, and the residue was separated by medium pressure column chromatography over silica gel using hexane-benzene as an eluent. Two types of 2:1-adducts **5-A** and **5-B** were obtained together with 1,3-oxathiole-2-thione derivative **6**.

Major 2:1-Adduct 5a-A: Yield 64.5% from **1a**; colorless crystals; mp 236.1—237.0 °C (from benzene-hexane); IR (KBr) 1697 (C=O), 1597, 1456, 1308, 1292, 1272, 1237, 1206, 1149, 1097, 1075, 1054, 1042, 1022, 968, 846, 756, 740, and

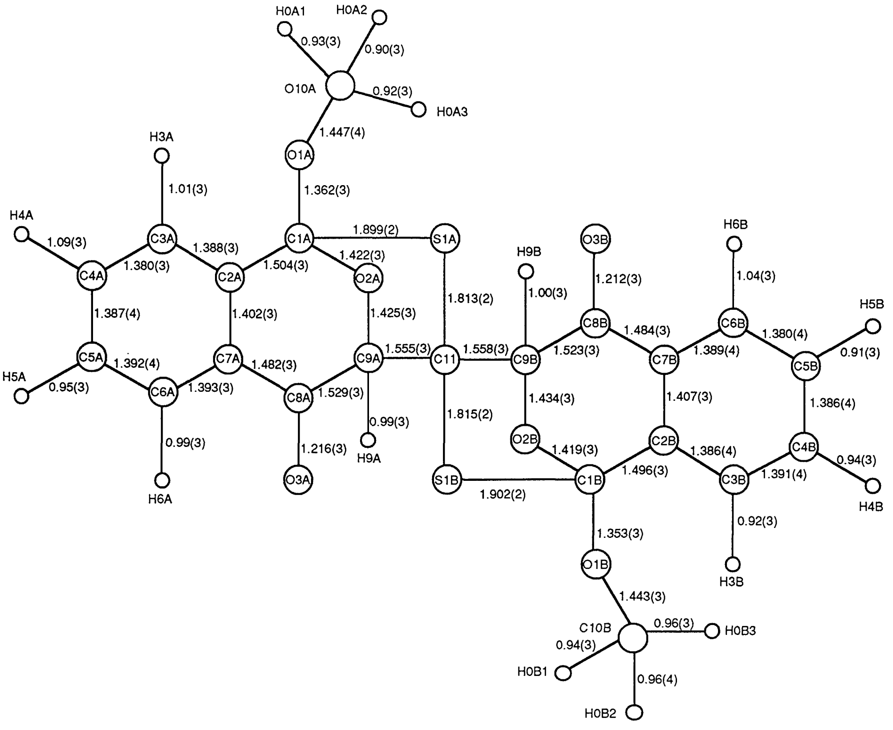
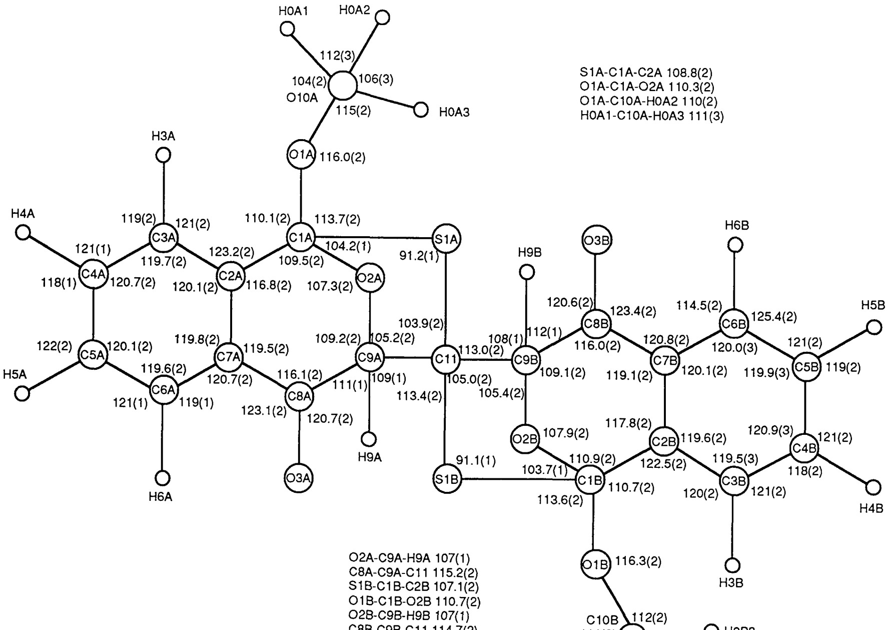


Fig. 4-a. Bond lengths of **5a-A**.



S1A-C1A-C2A 108.8(2)
O1A-C1A-O2A 110.3(2)
O1A-C10A-H0A2 110(2)
H0A1-C10A-H0A3 111(3)

O2A-C9A-H9A 107(1)
C8A-C9A-C11 115.2(2)
S1B-C1B-C2B 107.1(2)
O1B-C1B-O2B 110.7(2)
O2B-C9B-H9B 107(1)
C8B-C9B-C11 114.7(2)
O1B-C10B-H0B2 112(2)
H0B1-C10B-H0B3 102(3)
S1A-C11-S1B 113.5(1)
C9A-C11-C9B 108.1(2)

Fig. 4-b. Bond angles of **5a-A**.

656 cm^{-1}; MS (EI) 328 (1.4%, M$^+$), 396 (8.4), 176 (100), 133 (8), and 77 (10). Found: C, 58.93; H, 3.77%. Calcd for $C_{21}H_{16}O_6S_2$: C, 58.87; H, 3.76%.

Minor 2:1-Adduct 5a-B: Yield 10.3% from **1a**; colorless crystals; mp 200—202 °C (from benzene-hexane); IR (KBr) 2985, 2942, 1700 (C=O), 1598, 1455, 1435, 1318, 1291, 1267, 1242, 1202, 1146, 1085, 1058, 1021, 1009, 980, 962, 904, 849, 794, 765, 852, 711, and 655 cm^{-1}; MS (EI) 423 (3.4%, M$^+$), 396 (8.4), 176 (3), 163 (100), 133 (8), and 77 (8).

5-(*o*-Methoxycarbonylphenyl)-1,3-oxathiole-2-thione (6a):

Yield 6.3% from **1a**; pale yellow crystals; mp 120.1—121.0 °C (from benzene–hexane).[7]

Major 2:1-Adduct 5b-A: Yield 65.7% from **1b**; colorless crystals; mp 211.0—211.8 °C (from acetone); IR (KBr) 3070, 2977, 1700 (C=O), 1598, 1456, 1384, 1371, 1294, 1270, 1236, 1190, 1171, 1140, 1095, 1074, 1053, 998, 968, 914, 894, 885, 834, 807, 775, 758, 747, 688, and 653 cm^{-1}. MS (EI) 484 (3%, M^+), 452 (7), 424 (6), 382 (14), 367 (7), 349 (6), 323 (7), 294 (9), 252 (7), 234 (19), 191 (17), 149 (100), 133 (7), 111 (6), and 104 (8). Found: C, 61.87; H, 5.23%. Calcd for $C_{25}H_{24}O_6S_2$: C, 61.96; H, 4.99%.

Minor 2:1-Adduct 5b-B: Yield 8.8% from **1b**; colorless crystals; mp 210.8—211.2 °C (from benzene–hexane); IR (KBr) 3191, 3073, 2975, 1697 (C=O), 1598, 1457, 1383, 1371, 1292, 1269, 1243, 1191, 1171, 1141, 1111, 1099, 1079, 1056, 993, 926, 903, 854, 836, 792, 753, 720, 701, 669, and 653 cm^{-1}; MS (EI) 484 (8%, M^+), 452 (5), 424 (10), 382 (7), 377 (7), 349 (5), 294 (10), 251 (8), 234 (19), 191 (13), 162 (6), 149 (100), 133 (5), 111 (4), and 104 (11). Found: C, 62.16; H, 5.04%. Calcd for $C_{25}H_{24}O_6S_2$: C, 61.96; H, 4.99%.

5-(*o*-Isopropoxycarbonylphenyl)-1,3-oxathiole-2-thione (6b): Yield 8.8% from **1b**; yellow oil.[7]

X-Ray Crystallography of the 2:1-Adduct 5a-A. Single crystals of **5a-A** recrystallized from a mixed solvent of benzene–hexane were used for X-ray analysis. Intensity data were measured on a Rigaku four-circle diffractometer with Ni-filtered Cu $K\alpha(\lambda=1.5418$ Å) radiation. The reflections of 2752 with $|F_o|\geq 2\sigma(F_o)$ were used in the structure determination. The space group $P\bar{1}$ was confirmed by the statistical distribution of E values and a successful refinement of the structure.

The crystal structure was determined by direct methods with the program of MULTAN 80.[12] The E-map computed with the phase set of the highest combined figure of merit (2.864) revealed the positions of all the non-hydrogen atoms. The hydrogen atoms were located on a difference map and their positional parameters were refined together with isotropic thermal parameters, while the other atoms were refined anisotropically by block-diagonal least-squares. The final R value was 0.044. The function minimized was $\sum w(|F_o|-k|F_c|)^2$ with the following weighting scheme: $w=1/(\sigma^2(F_o)+0.0476|F_o|+0.0041|F_o|^2)$.

All the atomic scattering factors were taken from the "International Tables for X-Ray Crystallography".[14] The computations were carried out with "The Universal Crystallographic Computing System-Osaka"[15] on an ACOS-S930 computer at the Research Center for Protein Engineering, Institute for Protein Research, Osaka University. Table 3 lists the final atomic and thermal parameters with their estimated standard deviations. The bond distances and angles are given in Fig. 4.[16]

Table 3. Atomic Positional and Thermal Parameters for the Non-Hydrogen Atoms of **5a-A**

Atom	x	y	z	$B_{eq}/Å^2$
S(1A)	0.87328(4)	0.26793(4)	0.38567(8)	3.18
S(1B)	0.63559(5)	0.18057(5)	0.49898(9)	4.00
O(1A)	0.9683(1)	0.2391(1)	0.0732(2)	3.81
O(2A)	0.7647(1)	0.1583(1)	0.0244(2)	3.11
O(3A)	0.5752(1)	−0.0769(1)	0.2076(3)	4.21
O(1B)	0.4266(1)	0.2271(1)	0.4372(2)	3.77
O(2B)	0.5483(1)	0.2938(1)	0.2372(2)	3.50
O(3B)	0.8233(2)	0.5226(2)	0.2867(3)	5.32
C(1A)	0.8776(2)	0.1791(2)	0.1555(3)	2.88
C(2A)	0.8870(2)	0.0602(2)	0.2033(3)	2.77
C(3A)	0.9934(2)	0.0372(2)	0.2296(3)	3.22
C(4A)	0.9944(2)	−0.0749(2)	0.2710(3)	3.69
C(5A)	0.8904(2)	−0.1651(2)	0.2875(4)	3.80
C(6A)	0.7840(2)	−0.1423(2)	0.2648(3)	3.51
C(7A)	0.7821(2)	−0.0293(2)	0.2231(3)	2.92
C(8A)	0.6693(2)	−0.0039(2)	0.1926(3)	3.00
C(9A)	0.6757(2)	0.1196(2)	0.1325(3)	3.05
C(10A)	0.9622(2)	0.3486(2)	−0.0017(5)	5.31
C(1B)	0.5419(2)	0.2783(2)	0.4271(3)	3.19
C(2B)	0.6022(2)	0.3963(2)	0.5600(3)	3.31
C(3B)	0.5657(2)	0.4219(2)	0.7261(4)	4.01
C(4B)	0.6279(2)	0.5325(2)	0.8454(4)	4.81
C(5B)	0.7263(2)	0.6168(2)	0.8022(4)	4.78
C(6B)	0.7626(2)	0.5923(2)	0.6364(4)	4.37
C(7B)	0.7016(2)	0.4823(2)	0.5151(3)	3.54
C(8B)	0.7419(2)	0.4521(2)	0.3403(4)	3.66
C(9B)	0.6715(2)	0.3270(2)	0.2242(3)	3.38
C(10B)	0.3571(2)	0.1141(2)	0.3106(4)	4.66
C(11)	0.7117(2)	0.2239(2)	0.3046(3)	3.05

$B_{eq}=(4/3)\{a^2B_{11}+b^2B_{22}+c^2B_{33}+ab\cos\gamma(B_{12}/2)+ac\cos\beta(B_{13}/2)+bc\cos\alpha(B_{23}/2)\}$.

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