Reactions of *N*,*N*-(dialkyl)arylthioacetamides with dialkyl acetylenedicarboxylates

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2-(Alkoxycarbonylmethylidene)-4-aryl-5-(dialkylamino)thiophen-3(2*H*)-ones were synthesized by condensation of *N*,*N*-(dialkyl)arylthioacetamides with dialkyl acetylenedicarboxylates. Intermediate substituted vinylic sulfides were isolated. When heated or in the presence of an acid or a base, they undergo cyclization into thiophenes.

Key words: thiophene, *N*,*N*-(dialkyl)arylthioacetamides, dialkyl acetylenedicarboxylates, condensation.

The reactions of thioureas, thiosemicarbazides, and thioamides with acetylenedicarboxylic acid derivatives are convenient methods for the synthesis of nitrogen- and sulfur-containing heterocycles[1—9] such as thiazoles,[1—5] thiazines,[6,7] imidazoles,[8] thiadiazines, and pyrimidines.[9] However, only nonsubstituted or N-monosubstituted derivatives of thioureas, thiosemicarbazides, and thioamides were involved in this reaction. The goal of the present study is to investigate the reactions of *N*,*N*-(dialkyl)arylthioacetamides with dialkyl acetylenedicarboxylates.

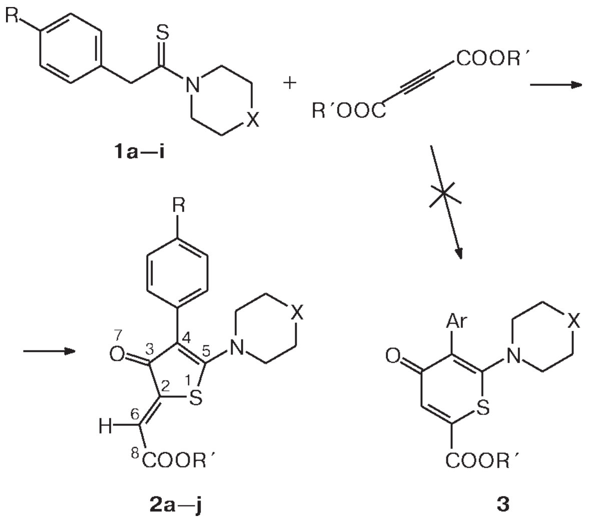
Results and Discussion

The reactions of 2-arylthioacetamides **1a—i** with acetylene derivatives yielded only substituted 2-methylidenethiophen-3-ones **2a—j** (Scheme 1), which were identified using IR and ^{1}H and ^{13}C NMR spectroscopy and mass spectrometry (Tables 1—3).

^{13}C NMR data enable one to exclude the formation of possible products **3**. Thus, a signal from the C(4) atom shows a splitting only for the *ortho*-hydrogen atoms of the phenyl group, and $J_{C(3)-H(6)} = 4.6$ Hz, which indicates the (*Z*)-configuration of the exocyclic double bond, $J_{C(8)-H(6)}$ being equal to 1.9 Hz. The structure of compound **2j** was confirmed by X-ray diffraction data for a single crystal obtained by crystallization from ethanol.

Two crystallographically independent molecules of compound **2j** are very close in geometry. Their thiophene fragments are planar with an accuracy of ±0.016(2) Å, and the whole molecule, except for the phenyl and piperidine groups, is planar with an accuracy of ±0.228(6) Å.

Scheme 1



Compound	1, 2 X	1, 2 R	2 R´	Compound	1, 2 X	1, 2 R	2 R´
1a, **2a**	O	OMe	Me	**1f**, **2f**	O	OEt	Et
1b, **2b**	O	Cl	Me	**1g**, **2g**	CH_2	H	Me
1c, **2c**	O	Br	Me	**1h**, **2h**	CH_2	OMe	Me
1d, **2d**	O	H	Me	**1i**, **2i**	CH_2	Cl	Me
1e, **2e**	O	Me	Et	**2j**	CH_2	H	Et

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Table 1. Characteristics of the starting thioacetamides **1** and **4**

Compound	M.p./°C	Yield (%)	Found/Calculated (%)		Molecular formula	MS, $[M]^+$
			N	S		
1a	70	69	—	13.08 12.76	$C_{13}H_{17}NO_2S$	251
1b	85	48	—	12.81 12.54	$C_{12}H_{14}ClNOS$	255
1c	70	52	—	10.89 10.68	$C_{12}H_{14}BrNOS$	301, 299
1d	100	38	—	14.86 14.49	$C_{12}H_{15}NOS$	221
1e	105	75	—	13.85 13.63	$C_{13}H_{17}NOS$	235
1f	80	47	—	12.28 12.08	$C_{14}H_{19}NO_2S$	265
1g	80	65	—	15.08 14.62	$C_{13}H_{17}NS$	219
1h	70	36	—	12.98 12.86	$C_{14}H_{19}NOS$	249
1i	60	57	—	13.01 12.63	$C_{13}H_{16}ClNS$	253
4a	95	69	16.55 16.46	18.69 18.84	$C_7H_{10}N_2OS$	170
4b	65	38	16.41 16.65	18.95 19.06	$C_8H_{12}N_2S$	168
4c	100	55	18.32 18.16	21.12 20.79	$C_7H_{10}N_2S$	154

Table 2. Characteristics of compounds **2**, **5**, and **6**

Compound	M.p./°C	Yield (%)	Found/Calculated (%)		Molecular formula	MS, $[M]^+$
			N	S		
2a	160	45	4.12 3.88	8.97 8.86	$C_{18}H_{19}NO_5S$	361
2b	215	51	3.98 3.83	9.08 8.76	$C_{17}H_{16}ClNO_4S$	365
2c	220	62	3.74 3.41	8.11 7.81	$C_{17}H_{16}BrNO_4S$	410
2d	150	42	4.43 4.23	9.39 9.68	$C_{17}H_{17}NO_4S$	331
2e	220	54	3.63 3.90	9.35 8.92	$C_{19}H_{21}NO_4S$	359
2f	145	47	3.62 3.60	8.14 8.23	$C_{20}H_{23}NO_5S$	389
2g	140	56	4.10 4.25	9.89 9.73	$C_{18}H_{19}N0_4S$	329
2h	155	52	4.25 3.90	8.77 8.92	$C_{19}H_{21}N0_4S$	359
2i	185	48	4.13 4.00	10.05 9.17	$C_{17}H_{16}ClNO_3S$	349
2j	141	53	3.90 4.08	8.89 9.33	$C_{19}H_{21}NO_3S$	343
5a	165	57	10.15 9.99	11.26 11.44	$C_{12}H_{12}N_2O_4S$	280

(*to be continued*)

Table 2 (*continued*)

Compound	M.p./°C	Yield (%)	Found/Calculated (%)		Molecular formula	MS, $[M]^+$
			N	S		
5b	180	60	10.21 10.06	11.29 11.52	$C_{13}H_{14}N_2O_3S$	278
5c	240	53	10.52 10.60	11.82 12.13	$C_{12}H_{12}N_2O_3S$	264
5d	238	85	10.25 10.06	11.78 11.52	$C_{13}H_{14}N_2O_3S$	278
6a	115	62	9.43 8.97	10.52 10.25	$C_{13}H_{16}N_2O_5S$	312
6d	96	65	9.73 9.45	11.22 10.82	$C_{13}H_{16}N_2O_4S$	296

Table 3. ^{1}H NMR spectra of the compounds synthesized

Compound	^{1}H NMR (DMSO-d_6) δ (*J*/Hz)
2a	7.16 (d, 2 H, ArH, *J* = 6.8); 6.93 (d, 2 H, ArH, *J* = 6.8); 6.63 (s, 1 H, CH); 3.77 (s, 3 H, OMe); 3.76 (s, 3 H, OMe), 3.60—3.64 (m, 4 H, 2 CH_2); 3.44—3.48 (m, 4 H, 2 CH_2)
2b	7.42 (d, 2 H, ArH, *J* = 6.7); 7.29 (d, 2 H, ArH, *J* = 6.7); 6.66 (s, 1 H, CH); 3.78 (s, 3 H, OMe); 3.60—3.64 (m, 4 H, 2 CH_2); 3.44—3.48 (m, 4 H, 2 CH_2)
2c	7.52—7.56 (m, 2 H, ArH); 7.21—7.24 (m, 2 H, ArH); 6.65 (s, 1 H, CH); 3.77 (s, 3 H, OMe); 3.60—3.66 (m, 4 H, 2 CH_2); 3.41—3.46 (m, 4 H, 2 CH_2)
2d	7.30—7.32 (m, 5 H, C_6H_5); 6.65 (s, 1 H, CH); 3.79 (s, 3 H, OMe); 3.60—3.65 (m, 4 H, 2 CH_2); 3.40—3.45 (m, 4 H, 2 CH_2)
2e	7.10—7.16 (m, 4 H, ArH); 6.61 (s, 1 H, CH); 4.24 (q, 2 H, CH_2, *J* = 7.1); 3.58—3.62 (m, 4 H, 2 CH_2); 3.42—3.46 (m, 4 H, 2 CH_2); 2.30 (s, 3 H, Me); 1.28 (t, 3 H, Me, *J* = 7.1)
2f	7.10 (d, 2 H, ArH, *J* = 7.8); 6.84 (d, 2 H, ArH, *J* = 7.8); 6.63 (s, 1 H, CH); 4.25 (q, 2 H, CH_2, *J* = 7.0); 4.01 (q, 2 H, CH_2, *J* = 7.0); 3.60—3.64 (m, 4 H, 2 CH_2); 3.28—3.33 (m, 4 H, 2 CH_2); 1.38 (t, 3 H, Me, *J* = 7.0); 1.26 (t, 3 H, Me, *J* = 7.0)
2g	7.30—7.40 (m, 5 H, C_6H_5); 6.63 (s, 1 H, CH); 3.76 (s, 3 H, OMe); 3.40—3.44 (m, 4 H, $N(CH_2)_2$); 1.50—1.60 (m, 6 H, $(CH_2)_3$)
2h	7.16 (d, 2 H, ArH, *J* = 8.9); 6.93 (d, 2 H, Ar, *J* = 8.9); 6.61 (s, 1 H, CH); 3.77 (s, 3 H, OMe); 3.76 (3 H, s, OMe); 3.43 (br.s, 4 H, $N(CH_2)_5$); 1.56 (br.s, 6 H, $N(CH_2)_5$)
2i	7.42 (d, 2 H, ArH, *J* = 7.9); 7.28 (d, 2 H, ArH, *J* = 7.9); 6.62 (s, 1 H, CH); 3.77 (s, 3 H, OMe); 3.40—3.44 (m, 4 H, $N(CH_2)_2$); 1.55—1.60 (m, 4 H, $(CH_2)_3$)
2j*	7.28—7.33 (m, 2 H, C_6H_5); 6.20—6.30 (m, 3 H, C_6H_5); 6.84 (s, 1 H, CH); 4.25 (t, 2 H, OCH_2, *J* = 7.0); 3.73 (t, 4 H, 2 NCH_2); 1.53 (tt, 4 H, 2 CH_2); 1.59 (tt, 2 H, CH_2); 1.30 (q, 3 H, Me, *J* = 7.0)
5a	6.70 (s, 1 H, CH); 3.80—3.84 (m, 4 H, $O(CH_2)_2$); 3.75 (s, 3 H, OMe); 1.30—1.35 (s, 4 H, $N(CH_2)_2$)
5b	6.69 (s, 1 H, CH); 4.16—4.20 (m, 2 H, CH_2); 3.80 (s, 3 H, OMe); 3.70—3.76 (m, 2 H, CH_2); 1.75—1.81 (m, 6 H, $(CH_2)_3$)
5c	6.68 (s, 1 H, CH); 4.00—4.10 (m, 2 H, CH_2); 3.80 (s, 3 H, OMe); 3.75—3.79 (m, 2 H, CH_2); 2.00—2.15 (4 H, m, $(CH_2)_2$)
5d	6.67 (s, 1 H, CH); 4.20 (q, 2 H, OCH_2, *J* = 7.1); 4.00—4.10 (m, 2 H, CH_2); 3.75—3.79 (m, 2 H, CH_2); 2.00—2.15 (m, 4 H, $(CH_2)_2$); 1.28 (q, 3 H, Me, *J* = 7.1)
6a	6.72 (s, 1 H, CH); 4.85 (s, 1 H, CH); 3.70 (s, 3 H, OMe); 3.74 (s, 3 H, OMe); 3.50—3.58 (m, 4 H, $O(CH_2)_2$); 3.15—3.19 (m, 4 H, $N(CH_2)_2$)
6d*	Major product (90%): 6.72 (s, 1 H, CH); 4.36 (s, 1 H, CH); 4.21 (q, 2 H, $O\underline{CH_2}Me$, *J* = 7.1); 4.20 (q, 2 H, $O\underline{CH_2}Me$, *J* = 7.1); 3.56—3.66 (m, 4 H, 2 CH_2); 3.24 (m, 4 H, 2 CH_2); 1.30 (t, 3 H, Me, *J* = 7.1); 1.28 (t, 3 H, Me, *J* = 7.1) Minor product (10%): 6.56 (s, 1 H, CH); 4.32 (s, 1 H, CH); 4.21 (q, 2 H, $O\underline{CH_2}Me$, *J* = 7.1); 4.20 (q, 2 H, $O\underline{CH_2}Me$, *J* = 7.1); 3.56—3.66 (m, 4 H, 2 CH_2); 3.24 (m, 4 H, 2 CH_2); 1.30 (t, 3 H, Me, *J* = 7.1); 1.28 (t, 3 H, Me, *J* = 7.1)

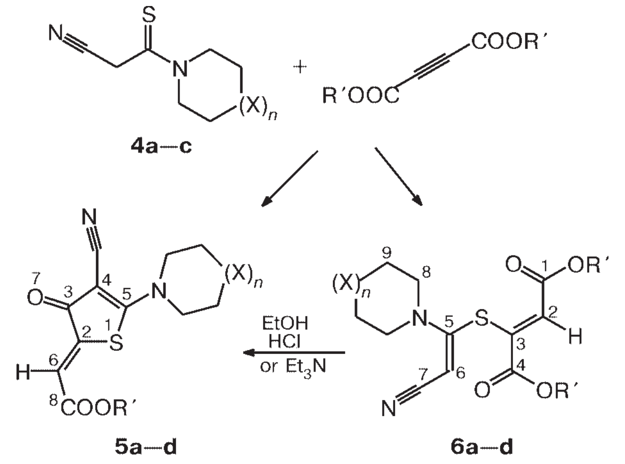
* In $CDCl_3$.

It should be noted that the bond lengths in the C(5)=C(4)—C(3)=O(7) fragment are equalized (the average bond lengths for two molecules are 1.389, 1.431, and 1.226 Å, respectively) and that the S(1)—C(2) and S(1)—C(5) bonds are not equivalent (average bond lengths 1.736 and 1.796 Å). The above structural features were confirmed by MP2/6-31G* quantum-chemical calculations for (5-amino-3-oxo-2*H*-thiophen-2-ylidene)acetic acid as a model compound; bond equalization in the C(5)=C(4)—C(3)=O(7) fragment was noted earlier for structurally close compounds.[10,11]

Unlike compounds **1a—i**, cyanothioacetamides **4a—c** react with dialkyl acetylenedicarboxylates to give not only thiophenes **5a—d**, but also vinylic sulfides **6a,d** resulting from the addition of thioamides to acetylene derivatives (Scheme 2).

Sulfide **6d** contains an impurity (~10%), which is probably a stereoisomer about one of the double bonds. When heated in ethanol, compounds **6a,d** undergo cyclization into thiophenes **5a,d**. This reaction is also possible at room temperature in the presence of a catalytic amount of triethylamine or HCl. The structures of all the compounds obtained were confirmed by IR, ^{1}H and ^{13}C NMR, and MS data. As in the case of aryl derivatives **2a—j**, the ^{13}C NMR spectra of compounds **5a—d** show the coupling constant $J_{C(3)-H(6)}$ = 4.6 Hz, which indicates (*Z*)-configuration of the exocyclic double bond.

Scheme 2



R′ = Me (**a—c**), CH$_2$Me (**d**)
X = O (**a**), CH$_2$ (**b**)
n = 1 (**a**, **b**), 0 (**c**, **d**)

Hence, *N*,*N*-(dialkyl)aryl- and -cyanothioacetamides react with dialkyl acetylenedicarboxylates to give substituted 5-methylidenethiophen-4-ones. At the first stage, acetylene derivatives add to the sulfur atom to form substituted vinylic sulfides, which undergo cyclization into thiophenones in the presence of an acid or a base or on heating.

Table 4. ^{13}C NMR spectra (CDCl$_3$) of the compounds synthesized

Compound	δ (*J*/Hz)
2d	184.84 (d, C(3), J = 4.5); 169.32 (d, C(5), J = 3.2); 167.15 (qd, C(8), J = 3.9, J = 1.7); 146.11 (d, C(2), J = 2.2); 133.81 (s, Ph); 129.89 (dd, Ph, J = 168.5, J = 4.5); 128.40 (dd, Ph, J = 169.8, J = 4.8); 127.24 (d, Ph, J = 171.2); 114.69 (d, C(6), J = 172.0); 108.39 (d, C(4), J = 3.9); 66.38 (tt, OCH$_2$, J = 148.0, J = 4.5); 52.19 (t, OMe, J = 147.4); 50.94 (tt, NCH$_2$, J = 148.0, J = 4.5)
2j	184.46 (d, C(3), J = 4.6); 169.42 (d, C(5), J = 4.0); 166.84 (td, C(8), J = 2.9, J = 1.9); 146.18 (d, C(2), J = 2.2); 134.58 (s, Ph); 129.79 (dd, Ph, J = 169.2, J = 4.5); 128.17 (dd, Ph, J = 168.5, J = 4.5); 126.78 (d, Ph, J = 170.1); 114.18 (d, C(6), J = 171.3); 107.65 (d, C(4), J = 3.6); 61.03 (tq, $O\underline{CH}_2Me$, J = 147.8, J = 4.6); 52.22 (tt, NCH$_2$); 25.83 (tt, CH$_2$); 23.56 (t, CH$_2$, J = 147.5); 14.01 (qt, OCH_2CH_3, J = 127.2, J = 2.4)
5d	183.54 (d, C(3), J = 5.0); 165.90 (d, C(5), J = 2.4); 172.04 (t, C(8), J = 3.5); 143.48 (d, C(2), J = 2.2); 116.19 (d, C(6), J = 172.2); 115.20 (s, CN); 79.16 (s, C(4)); 61.51 (tq, $O\underline{CH}_2Me$, J = 148.1, J = 4.5); 53.68 (t, NCH$_2$); 51.85 (t, NCH$_2$); 26.13 (t, CH$_2$); 25.83 (t, CH$_2$); 23.11 (t, CH$_2$); 13.80 (qt, $OCH_2\underline{CH}_3$ J = 127.4, J = 2.6)
6d	Major product (90%): 164.31 (dt, C(1), J = 3.2, J = 3.2); 163.34 (dt, C(4), J = 6.0, J = 3.0); 160.44 (br.s, C(5), $W_{1/2}$ = 6); 142.96 (s, C(3)); 125.54 (d, C(2), J = 169.6); 118.06 (s, C(7)); 75.82 (d, C(6), J = 169.4); 65.70 (tm, C(9), J = 144.4); 62.62 (tq, $O\underline{CH}_2CH_3$, J = 148.5, J = 4.5); 61.29 (tq, $O\underline{CH}_2Me$ J = 148.6, J = 4.5); 49.63 (tm, C(8), J = 138.4); 13.94 (qt, $OCH_2\underline{CH}_3$, J = 127.4, J = 2.6); 13.85 (qt, $OCH_2\underline{CH}_3$, J = 127.4, J = 2.6) Minor product (10%): 164.35 (C(1)); 163.52 (C(4)); 161.76 (C(5)); 143.61 (C(3)); 125.51 (C(2)); 117.94 (s, C(7)); 71.28 (d, C(6), J = 177.4); 66.34 (tm, C(9), J = 145.2); 62.90 ($O\underline{CH}_2CH_3$); 61.37 ($O\underline{CH}_2Me$); 51.01 (t, C(8), J = 145.2); 13.98 ($\underline{CH}_3$); 13.82 (CH$_3$)

Experimental

^{1}H and ^{13}C NMR spectra were recorded at 30 °C on Bruker 250 (250.13 MHz for ^{1}H) and Bruker DRX-500 instruments (500.13 and 125.13 MHz, respectively). IR spectra were recorded on a IR-75 spectrometer (in pellets with KBr). The course of the reactions was monitored and the purity of the products was checked by TLC on Silufol UV-254 plates in ethyl acetate—hexane (1 : 1), chloroform—ethanol (9 : 1), and chloroform. Melting points are given noncorrected. Solvents were purified and dried according to the standard procedures; THF was pre-dried over NaOH, distilled over Na, and kept over 4A molecular sieves. The physicochemical characteristics of the compounds obtained are given in Tables 1—3.

Synthesis of arylthioacetamides 1a—i (general procedure). A mixture of the corresponding acetophenone (0.027 mol), sulfur (1.3 g, 0.041 mol), and a cyclic amine (0.41 mol) was refluxed for 20 h. The resulting dark brown mixture was cooled and diluted with 100 mL of cold water. Trituration by a glass rod gave a crystalline precipitate. Thioacetamide **1a—i** was filtered off and recrystallized from ethanol.

Synthesis of cyanothioacetamides 4a—c (general procedure). Finely divided P_2S_5 (1.0 g) was added to a stirred suspension of the corresponding cyanoacetamide (2.0 g) in 100 mL of THF. The reaction mixture was heated at 50 °C for 30 min, cooled, and concentrated at a reduced pressure. Water (100 mL) was added, and the cyanothioacetamide obtained was recrystallized with coal and dried over P_2O_5.

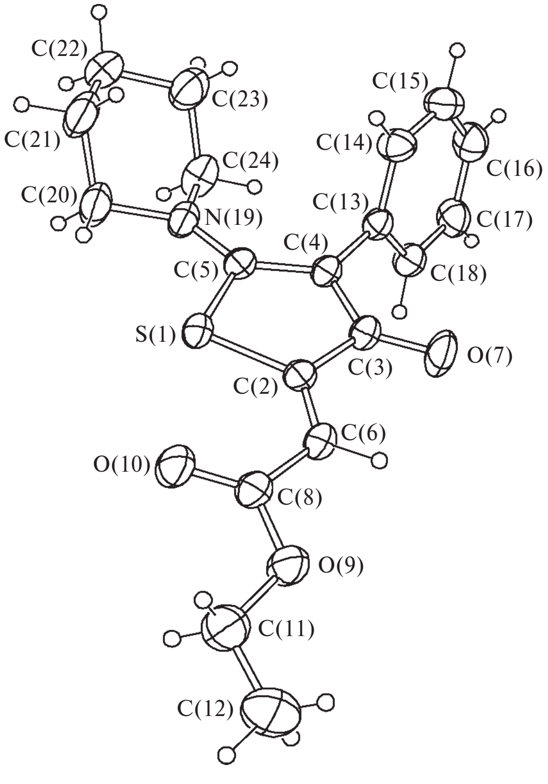


Fig. 1. Structure of one of the two crystallographically independent molecules of compound **2j** (according to the X-ray diffraction data).

Table 5. Selected bond lengths (*d*) in structure **2j**

Bond	*d*/Å		
	2j (X-ray diffraction analysis)*		Model compound**
	A	B	
S(1)—C(5)	1.799(4)	1.793(4)	1.786
S(1)—C(2)	1.736(4)	1.736(4)	1.747
C(4)=C(5)	1.397(6)	1.380(6)	1.364
C(3)—C(4)	1.425(6)	1.437(6)	1.440
C(3)=O(7)	1.225(5)	1.227(5)	1.234
C(5)—N(19)	1.323(6)	1.339(6)	1.352
C(2)—C(3)	1.509(6)	1.504(6)	1.521

* Two crystallographically independent molecules A and B.

** Calculated by the PM3/6-31G* method.

Synthesis of thiophenes (general procedure). Dimethyl acetylenedicarboxylate (0.1 mol) was added to a solution of *N*,*N*-dialkylthioacetamide **1** or **4** (0.1 mol) in 10 mL of anhydrous EtOH. The reaction mixture was refluxed for 2 h and concentrated, and the precipitate that formed was triturated with EtOH and filtered off. The product was recrystallized from a chloroform—hexane mixture.

X-ray diffraction analysis of compound 2j was carried out on a Bruker P4 diffractometer (Mo-Kα radiation, graphite monochromator, θ/2θ scan mode, 2θ < 50°). Bright red crystals are monoclinic: $a = 11.175(2)$ Å, $b = 11.512(2)$ Å, $c = 28.101(5)$ Å, $\beta = 95.726(8)°$, $V = 3597.2(10)$ Å^3, space group *Cc*, $C_{19}H_{21}NO_3S$, $Z = 8$, molecular mass 343.43, $d_{\text{calc}} = 1.268$ g cm^{-3}, $\mu = 0.196$ mm^{-1}. The intensities of 3350 independent reflections were measured for a crystal 0.1×1.2×1.4 mm in size; corrections for crystal habit absorption were applied (transmission 0.84—0.97). The structure was solved by the direct method using the SHELXS-97 program and refined by the least-squares method in the anisotropic-isotropic approximation using the SHELXL-97 program to $wR_2 = 0.1303$; $S = 1.026$ for all reflections ($R = 0.0456$ for 2974 $F > 4\sigma$). Hydrogen atoms were located geometrically and refined isotropically with the fixed carbon atoms which they are bonded to. Structure **2j** is displayed in Fig. 1. Selected bond lengths are given in Table 5.

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