Heteroatom Chemistry
Volume 8, Number 1, 1997

Syntheses and Structures of New Substituted 4-Aryl-sulfonyl-4H-1,3,4-thiadiazin-5(6H)-ones

Guofeng Jia* and Zhengming Li

Elemento-Organic Chemistry Institute, Nankai University, Tianjin, China

Received 7 May 1996

ABSTRACT

The cyclization of 4-aryl-1-arylsulfonylthiosemicarbazides **3***, which were prepared by treatment of arylsulfonyl chlorides* **1** *with 4-aryl-3-thiosemicarbazides* **2***, with chloroacetyl chloride, provided the corresponding 2-arylamino-4-arylsulfonyl-4H-1,3,4-thiadiazin-5(6H)-ones* **4** *in good yields. The structures of all these compounds were evaluated by elemental analyses, and* ^{1}*HNMR, and IR spectroscopy. The structure of compound* **4f** *was ascertained by X-ray diffraction analysis. © 1997 John Wiley & Sons, Inc.*

INTRODUCTION

Sulfonylureas and heterocyclic sulfonamides are widely known for their unprecedented levels of herbicidal activity [1–4]. New sulfonamides **3a–i** and **4a–i** have now been designed and synthesized in our laboratory.

RESULTS AND DISCUSSION

The usual reaction reported for the syntheses of 4-aryl-1-arylsulfonylthiosemicarbazides is the addition of arylsulfonylhydrazines to aryl isothiocyanates [5–8]. Actually, treatment of arylsulfonyl chlorides **1** in the presence of pyridine with thiosemicarbazides **2** [9] afforded **3a–i** in good yields (Table 1). Physical properties and spectral data are recorded in Tables 1 and 2, respectively.

Cyclizations of compounds **3a–i** with chloroacetyl chloride were studied here. The reactions took place smoothly in an ice bath in the presence of triethylamine. We believed that the six-member ring compounds **4a–i** were the most likely products to be formed. However, earlier work has shown that the five-member ring compounds **5a–i** or **6a–i** could also be formed under some conditions [10]. Tables 1 and 2 list the physical properties and IR and ^{1}HNMR spectra of the products. Unfortunately, these data are insufficient to confirm which are the correct structures of the products, **4a–i**, *5a–i*, or **6a–i** (Scheme). This prompted us to carry out an X-ray diffraction analysis of one of the products.

Figure 1 depicts the molecular structure of compound **4f**. Tables 3, 4, and 5 list the geometrical features. These results indicate that the ring formed was indeed a six-member one. Also, the distances of C(1)–N(1) and C(1)–N(2) are 1.357 and 1.300 angstroms, respectively, and these indicate that C(1) and N(2) are connected by a double bond. Thus, the structure of compound **4f** was ascertained.

EXPERIMENTAL

Instruments

Melting points were determined on a micromelting-point apparatus and are uncorrected. IR spectra were recorded with a Shimadzo-IR 435 infrared spectrophotometer. ^{1}HNMR spectra were determined with either a Joel-FX-90Q(90MHz) or Bruker AC-P200(200MHz) spectrometer. Elemental analysis

---

*To whom correspondence should be addressed.

1042-7163/97/010071-06

TABLE 1 Physical Constants and Elemental Analyses of Compounds **3a–i** and **4a–i**

						Analysis (Calcd./Found)		
Compd.	*R*[1]	*R*[2]	*Yield %*	*Mp., °C*	*Formula*	*C*	*H*	*N*
3a[a]	4-CH_3	H	68	192.0–194.0	$C_{14}H_{15}N_3O_2S_2$	52.32	4.70	13.03
						52.66	4.64	12.88
3b[a]	4-CH_3	2-CH_3	84	195.0–195.8	$C_{15}H_{17}N_3O_2S_2$	53.71	5.11	12.53
						53.95	5.29	12.37
3c[a]	4-Cl	H	92	204.0–205.0 (dec.)	$C_{13}H_{12}ClN_3O_2S_2$	45.68	3.54	12.29
						45.86	3.51	12.24
3d	4-Cl	2-CH_3	86	202.0–202.5	$C_{14}H_{14}ClN_3O_2S_2$	47.25	3.96	11.82
						47.44	3.77	11.85
3e	4-Cl	3-Cl	82	195.0–197.0	$C_{13}H_{11}Cl_2N_3O_2S_2$	41.72	2.96	11.23
						41.73	2.78	10.99
3f	2-Cl	2-CH_3	68	182.0–182.5	$C_{14}H_{14}ClN_3O_2S_2$	47.25	3.96	11.81
						47.21	3.85	11.69
3g	4-NO_2	H	89	196.0–196.8 (dec.)	$C_{13}H_{12}N_4O_4S_2$	44.31	3.43	15.90
						44.34	3.48	15.54
3h	4-NO_2	2-CH_3	75	195.0–195.5	$C_{14}H_{14}N_4O_4S_2$	45.89	3.85	15.29
						45.61	3.86	15.46
3i	4-NO_2	3-Cl	83	180.5–181.0	$C_{13}H_{11}ClN_4O_4S_2$	40.36	2.87	14.48
						40.62	2.82	14.46
4a	4-CH_3	H	40	237.0–238.8	$C_{16}H_{15}N_3O_3S_2$	53.14	4.18	11.63
						53.33	4.17	11.69
4b	4-CH_3	2-CH_3	89	159.5–162.0	$C_{17}H_{17}N_3O_3S_2$	54.38	4.56	11.19
						54.57	4.68	11.05
4c	4-Cl	H	86	166.0–167.2	$C_{15}H_{12}ClN_3O_3S_2$	47.30	3.18	11.03
						47.15	3.08	11.25
4d	4-Cl	2-CH_3	82	155.5–156.5	$C_{16}H_{14}ClN_3O_3S_2$	48.54	3.56	10.61
						48.35	3.40	10.43
4e	4-Cl	3-Cl	83	187.5–189.5	$C_{15}H_{11}Cl_2N_3O_3S_2$	43.28	2.66	10.09
						43.50	2.64	9.92
4f	2-Cl	2-CH_3	85	165.0–165.8	$C_{16}H_{14}ClN_3O_3S_2$	48.54	3.56	10.61
						48.50	3.50	10.66
4g	4-NO_2	H	66	154.2–156.0	$C_{15}H_{12}N_4O_5S_2$	45.91	3.08	14.28
						45.55	3.18	13.88
4h	4-NO_2	2-CH_3	80	167.0–168.5	$C_{16}H_{14}N_4O_5S_2$	47.28	3.47	13.78
						47.37	3.32	13.45
4i	4-NO_2	3-Cl	75	187.8–189.0	$C_{15}H_{11}ClN_4O_5S_2$	42.21	2.60	12.12
						42.28	2.48	13.13

[a] Reported compounds [5].

data were obtained by use of a Yanaco CHN Corder MT-3 apparatus.

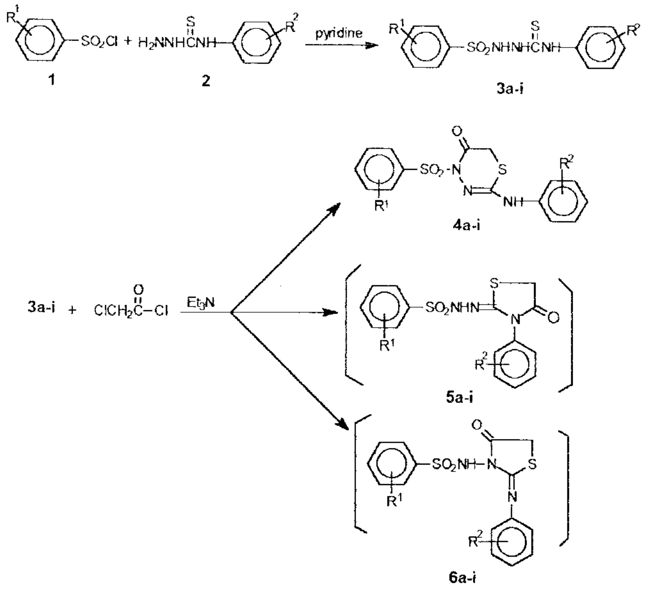
4-Aryl-1-arylsulfonylthiosemicarbazides (**3a–i**)*: General Procedure*

To a mixture of each 4-aryl-3-thiosemicarbazide **2** (0.015 mol), dry pyridine (0.017 mol), and anhydrous dichloromethane (25 mL) was added dropwise a solution of arylsulfonyl chloride (0.015 mol) in 25 mL of anhydrous dichloromethane, the temperature being maintained below 0°C. The reaction mixture was stirred for 8 hours at room temperature. After concentration of the reaction mixture, the residual solid was washed with cold water and recrystallized from acetone/petroleum ether. 4-Aryl-1-arylsulfonylthiosemicarbazides **3a–i** were thus obtained. All these compounds were characterized by elemental analyses and ^{1}HNMR and IR spectroscopy (Tables 1 and 2).

TABLE 2 ^{1}HNMR Data of Compounds **3a–i** and **4a–i**

		IR (cm^{-1}, KBr)			
Compd.	*^{1}HNMR (δ, deuterioacetone)*	N–H	C=S	C=O	SO_2
3a	2.43 (s, 3H, CH_3), 7.3–7.6 (m, 5H, Ph–H), 7.44 (d, 2H, *J* = 8.1) Hz, Ph–H), 7.82 (d, 2H, Ph–H), 8.82 (bs, NH), 8.92 (bs, NH), 9.44 (bs, NH).	3410w	1517s	1338m	1160m
3b	2.23 (s, 3H, CH_3), 2.46 (s, 3H, CH_3), 7.3–7.7 (m, 4H, Ph–H), 7.45 (d, 2H, *J* = 8.2 Hz, Ph–H), 7.83 (d, 2H, Ph–H), 8.81 (bs, NH), 8.94 (bs, NH), 9.21 (bs, NH).	3394w	1511s	1340s	1158m
3c	7.33–7.97 (m, 9H, Ph–H), 8.09 (bs, NH), 9.09 (bs, NH), 9.56 (bs, NH).	3402w	1519s	1349s	1174m
3d	2.24 (s, 3H, CH_3), 7.1–7.3 (m, 4H, Ph–H), 7.70 (d, 2H, *J* = 8.9 Hz, Ph–H), 7.94 (d, 2H, Ph–H), 8.95 (bs, NH), 9.13 (bs, NH), 9.29 (bs, NH).	3407w	1535s	1359s	1175m
3e	7.2–8.0 (m, 8H, Ph–H), 9.13 (bs, NH), 9.70 (bs, NH).	3354w	1532s	1346s	1163s
3f	2.27 (s, 3H, CH_3), 7.1–8.1 (m, 8H, Ph–H), 9.10 (bs, NH), 9.26 (bs, NH).	3403w	1538s	1337s	1166s
3g	7.1–7.4 (m, 5H, Ph–H), 6.09 (d, 2H, *J* = 8.0 Hz, Ph–H), 8.43 (d, 2H, Ph–H), 9.86 (bs, NH), 9.92 (bs, NH), 10.45 (bs, NH).	3401w	1529s	1346s	1166s
3h	2.14 (s, 3H, CH_3), 7.0–7.3 (m, 4H, Ph–H), 8.10 (d, 2H, *J* = 8.0 Hz, Ph–H), 8.46 (d, 2H, Ph–H), 9.80 (bs, NH), 10.50 (bs, NH).	3406w	1532s	1346s	1163s
3i	7.1–7.5 (m, 4H, Ph–H), 8.08 (d, 2H, *J* = 7.9 Hz, Ph–H), 8.45 (d, 2H, Ph–H), 10.02 (bs, NH), 10.07 (bs, NH), 10.49 (bs, NH).	3399w	1538s	1349s	1160s
4a	2.28 (s, 3H, CH_3), 2.04 (s, 2H, CH2), 6.48 (d, 2H, *J* = 7.2 Hz, Ph–H), 7.76 (d, 2H, Ph–H), 6.9–7.4 (m, 5H, Ph–H), 10.69 (bs, NH).	3407w	1757s	1339s	1159s
4b	[a] 2.29 (s, 3H, CH_3), 2.42 (s, 3H, CH_3), 3.38 (s, 2H, CH_3), 7.1–8.0 (m, 8H, Ph–H), 6.25 (bs, NH).	3409m	1691s	1357m	1185m
4c	[a] 3.37 (s, 2H, CH_2), 6.50 (bs, NH), 7.1–7.6 (m, 5H, Ph–H), 7.50 (d, 2H, *J* = 12.5 Hz, Ph–H), 8.01 (d, 2H, Ph–H).	3393m	1688s	1389m	1176s
4d	[a] 2.27 (s, 3H, CH_3), 3.37 (s, 2H, CH_2), 7.2–7.9 (m, 4H, Ph–H), 7.42 (d, 2H, *J* = 8.6 Hz, Ph–H), 7.97 (d, 2H, Ph–H).	3363m	1705s	1367m	1176s
4e	[a] 3.41 (s, 2H, CH_2), 7.1–7.8 (m, 4H, Ph–H), 7.48 (d, 2H, *J* = 8.7 Hz, Ph–H), 8.04 (d, 2H, Ph–H).	3386m	1707s	1369m	1163m
4f	[a] 2.26 (s, 3H, CH_3), 3.42 (s, 2H, CH_2), 7.0–7.6 (m, 4H, Ph–H), 7.30 (d, 2H, *J* = 8.0 Hz, Ph–H), 8.23 (d, 2H, Ph–H).	3411m	1704s	1361s	1182m
4g	2.08 (s, 3H, CH_3), 3.69 (s, 2H, CH_2), 6.7–7.8 (m, 5H, Ph–H), 8.32 (d, 2H, *J* = 8.6 Hz, Ph–H), 8.47 (d, 2H, Ph–H), 8.75 (bs, NH).	3340m	1704s	1361s	1182m
4h	2.31 (s, 3H, CH_3), 3.66 (s, 2H, CH_2), 7.0–7.7 (m, 4H, Ph–H), 8.17 (d, 2H, *J* = 8.9 Hz, Ph–H), 8.40 (d, 2H, Ph–H), 8.0 (bs, NH).	3390m	1707s	1344s	1174s
4i	3.72 (s, 2H, CH_2), 6.9–8.1 (m, 4H, Ph–H), 8.34 (d, 2H, *J* = 9.0 Hz, Ph–H), 8.48 (d, 2H, Ph–H), 8.94 (bs, NH).	3399m	1700s	1345m	1181s

[a]: $CDCl_3$.



R^1= 4-CH_3, 2-Cl, 4-Cl, 4-NO_2; R^2= H, 2-CH_3, 3-Cl

SCHEME 1

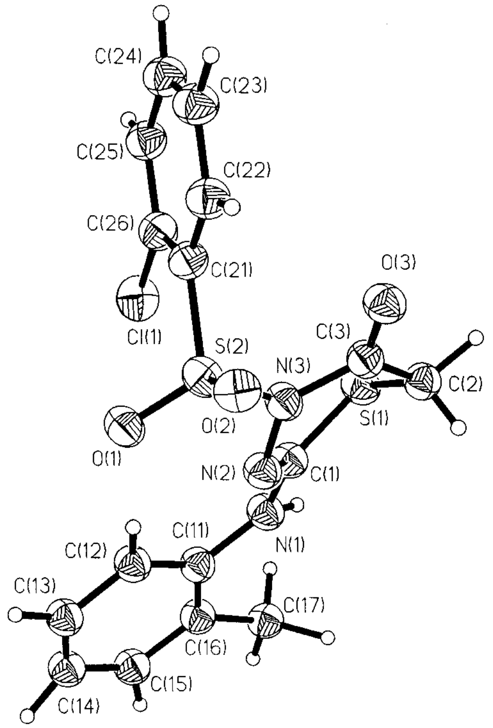


FIGURE 1 Molecular strucrure of **4f** as obtained from X-ray analysis.

TABLE 3 Positional Parameters and Their Estimated Standard Deviations of **4f**

Atom	x	y	z	*B(A²)ᵃ*
Cl(1)	1.0581(2)	0.0985(1)	1.2036(1)	4.50(2)
S(1)	1.2286(1)	0.1256(1)	1.5698(1)	3.59(2)
S(2)	1.2110(1)	−0.17537(9)	1.15065(9)	2.86(2)
O(1)	1.3010(4)	−0.0810(4)	1.0974(3)	3.98(6)
O(2)	1.2349(4)	−0.3140(3)	1.1168(4)	4.38(7)
O(3)	1.0812(4)	−0.2618(3)	1.3546(3)	3.75(6)
N(1)	1.4824(5)	0.2659(4)	1.5100(4)	3.82(8)
N(2)	1.3870(4)	0.0341(3)	1.3651(3)	2.85(6)
N(3)	1.2570(4)	−0.0880(3)	1.3204(3)	2.72(6)
C(1)	1.3741(5)	0.1377(4)	1.4691(4)	3.75(7)
C(2)	1.2200(5)	−0.0559(4)	1.5476(4)	3.36(7)
C(3)	1.1793(4)	−0.1492(4)	1.4000(4)	2.67(6)
C(11)	1.5884(5)	0.3240(4)	1.4388(4)	2.92(7)
C(12)	1.5774(5)	0.2611(4)	1.3021(4)	3.39(8)
C(13)	1.6821(6)	0.3261(5)	1.2380(5)	4.19(9)
C(14)	1.7943(7)	0.4536(5)	1.3113(6)	4.8(1)
C(15)	1.8052(6)	0.5148(5)	1.4467(6)	4.6(1)
C(16)	1.7028(6)	0.4515(4)	1.5154(5)	3.50(8)
C(17)	1.7217(8)	0.5198(5)	1.6659(6)	4.7(1)
C(21)	0.9996(5)	−0.1905(4)	1.1141(4)	3.16(8)
C(22)	0.8977(6)	−0.3235(5)	1.0640(6)	4.3(1)
C(23)	0.7328(7)	−0.3461(8)	1.0293(8)	5.7(2)
C(24)	0.6690(7)	−0.2290(9)	1.0505(6)	5.7(2)
C(25)	0.7691(6)	−0.0952(6)	1.1023(5)	4.7(1)
C(26)	0.9370(6)	−0.0737(5)	1.1343(4)	3.59(8)
H	1.473	0.307	1.584	5.0*
H1	1.750	0.445	1.723	5.0*
H2	1.622	0.543	1.686	5.0*
H3	1.808	0.604	1.698	5.0*
H4	1.305	−0.084	1.582	5.0*
H5	1.139	−0.084	1.609	5.0*
H12	1.500	0.168	1.250	5.0*
H13	1.668	0.279	1.141	5.0*
H14	1.891	0.500	1.279	5.0*
H15	1.891	0.584	1.500	5.0*
H22	0.918	−0.389	1.057	5.0*
H23	0.663	−0.440	0.991	4.0*
H24	0.557	−0.250	1.029	4.0*
H25	0.723	−0.027	1.111	4.0*

[a]Starred atoms were refined isotropically.

Isotropically refined atoms are given in the form of the isotropic equivalent displacement parameter defined as: (4/3) × [a2 × B (1, 1) + b2 × B (2, 2) + c2 × B (3, 3) + ab (cos gamma) × B (1, 2) + ac (cos beta) × B (1, 3) + bc (cos alpha) × B (2, 3)].

TABLE 4 Bond Distances in Angstroms of **4f**

Atom 1	*Atom 2*	*Distance*	*Atom 1*	*Atom 2*	*Distance*
C(1)	C(26)	1.737(2)	C(13)	C(14)	1.385(4)
S(1)	C(1)	1.750(2)	C(13)	H13	0.979(2)
S(1)	C(2)	1.810(3)	C(14)	C(15)	1.367(4)
S(2)	O(1)	1.434(2)	C(14)	H14	1.020(2)
S(2)	O(2)	1.429(2)	C(15)	C(16)	1.409
S(2)	N(3)	1.706(2)	C(15)	H15	0.894(2)
S(2)	C(21)	1.774(2)	C(16)	C(17)	1.510(4)
O(3)	C(3)	1.200(3)	C(17)	H1	1.187(3)
N(1)	C(1)	1.357(3)	C(17)	H2	0.966(3)
N(1)	C(11)	1.420(3)	C(17)	H3	0.956(2)
N(1)	H	0.789(2)	C(21)	C(22)	1.367(3)
N(2)	N(3)	1.406(2)	C(21)	C(26)	1.388(3)
N(2)	C(1)	1.300(3)	C(22)	C(23)	1.371(5)
N(3)	C(3)	1.380(2)	C(22)	H22	0.716(2)
C(2)	C(3)	1.516(3)	C(23)	C(24)	1.398(5)
C(2)	H4	0.924(2)	C(23)	H23	0.960(3)
C(2)	H5	1.077(2)	C(24)	C(25)	1.368(5)
C(11)	C(12)	1.380(3)	C(24)	H24	0.926(2)
C(11)	C(16)	1.399(3)	C(25)	C(26)	1.395(4)
C(12)	C(13)	1.393(3)	C(25)	H25	0.863(2)
C(12)	H12	0.991(2)			

Numbers in parentheses are estimated standard deviations in the least significant digits.

TABLE 5 Bond Angles in Degrees of **4f**

Atom 1	*Atom 2*	*Atom 3*	*Angle*	*Atom 1*	*Atom 2*	*Atom 3*	*Angle*
C(1)	S(1)	C(2)	94.55(8)	N(3)	C(3)	C(2)	113.6(1)
O(1)	S(2)	O(2)	118.8(1)	N(1)	C(11)	C(12)	122.4(2)
O(1)	S(2)	N(3)	105.10(8)	N(1)	C(11)	C(16)	115.8(2)
O(1)	S(2)	C(21)	110.9(2)	C(12)	C(11)	C(16)	121.8(2)
O(2)	S(2)	N(3)	110.1(1)	C(11)	C(12)	C(13)	119.5(2)
O(2)	S(2)	C(21)	108.5(1)	C(11)	C(12)	H12	120.8(2)
N(3)	S(2)	C(21)	102.30(8)	C(13)	C(12)	H12	119.8(3)
C(1)	N(1)	C(11)	129.9(2)	C(12)	C(13)	C(14)	120.0(2)
C(1)	N(1)	H	105.4(2)	C(12)	C(13)	H13	116.7(2)
C(11)	N(1)	H	124.6(3)	C(14)	C(13)	H13	123.4(2)
N(3)	N(2)	C(1)	115.6(1)	C(13)	C(14)	C(15)	120.3(2)
S(2)	N(3)	N(2)	114.5(1)	C(13)	C(14)	H14	126.1(3)
S(2)	N(3)	C(3)	118.7(1)	C(15)	C(14)	H14	112.1(2)
N(2)	N(3)	C(3)	126.4(2)	C(14)	C(15)	C(16)	121.4(3)
S(1)	C(1)	N(1)	114.9(1)	C(14)	C(15)	H15	122.6(3)
S(1)	C(1)	N(2)	125.0(1)	C(16)	C(15)	H15	114.3(3)
N(1)	C(1)	N(2)	120.1(1)	C(11)	C(16)	C(15)	117.1(3)
S(1)	C(2)	C(3)	110.8(1)	C(11)	C(16)	C(17)	122.8(3)
S(1)	C(2)	H4	123.7(2)	C(15)	C(16)	C(17)	120.3(3)
S(1)	C(2)	H5	106.8(1)	C(16)	C(17)	H1	113.0(2)
C(3)	C(2)	H4	104.0(2)	C(16)	C(17)	H2	108.4(2)
C(3)	C(2)	H5	116.4(2)	C(16)	C(17)	H3	108.8(3)
H4	C(2)	H5	95.2(1)	H1	C(17)	H2	108.3(3)
O(3)	C(3)	N(3)	122.6(2)	H1	C(17)	H3	109.0(2)
O(3)	C(3)	C(2)	123.7(2)	H2	C(17)	H3	109.4(2)
S(2)	C(21)	C(22)	117.3(2)	C(23)	C(24)	C(25)	120.7(2)
S(2)	C(21)	C(26)	122.5(2)	C(23)	C(24)	H24	114.9(3)
C(22)	C(21)	C(26)	120.3(3)	C(25)	C(24)	H24	124.4(4)
C(21)	C(22)	C(23)	121.5(2)	C(24)	C(25)	C(26)	120.3(2)
C(21)	C(22)	H22	127.9(3)	C(24)	C(25)	H25	116.3(3)
C(23)	C(22)	H22	110.4(3)	C(26)	C(25)	H25	123.3(3)
C(22)	C(23)	C(24)	118.4(3)	Cl(1)	C(26)	C(21)	122.8(2)
C(22)	C(23)	H23	120.8(3)	Cl(1)	C(26)	C(25)	118.4(2)
C(24)	C(23)	H23	120.8(3)	C(21)	C(26)	C(25)	118.8(2)

Numbers in parentheses are estimated standard deviations in the least significant digits.

2-Arylamino-4-arylsulfonyl-4H-1,3,4-thiadiazin-5(6H)-ones (**4a–i**)*: General Procedure*

Chloroacetyl chloride (0.005 mol) was added slowly at 0°C to a solution of each 2-aryl-1-arylsulfonylthiosemicarbazide **3a–i** (0.005 mol) and triethylamine (0.011 mol) in 30 mL of anhydrous tetrahydrofuran. After 30 minutes, the reaction mixture was allowed to warm to room temperature and was stirred for 6 hours, then concentrated in vacuo. The crude 2-arylamino-4-arylsulfonyl-4H-1,3,4-thiadiazin-5(6H)-ones **4a–i** were washed with cold water and recrystallized from acetone or chloroform. The elemental analyses and physical properties of these new compounds are given in Tables 1 and 2.

Single-Crystal X-ray Structure Determination of **4f**

Crystals suitable for X-ray diffraction analysis were obtained by recrystallization of compound **4f** from a mixture of chloroform and petroleum ether. The crystal used for data collection was a colorless prism having dimensions of 0.4 × 0.35 × 0.3 mm. Lattice constants and intensity data were measured at 299 K and $\lambda = 0.71073$ (M_0K). A total of 2539 reflections were collected, of which 2235 were unique. The space group was determined to be P-1. Cell data: a = 8.559 (2), b = 10.426 (1), c = 10.797 (2); $\alpha = 109.90\,(1)^0$, $\beta = 95.98\,(2)^0$, $\gamma = 101.89\,(1)^0$; $V = 870.5$ (6); $Z = 2$; $Dc = 1.51$ Mg/m^3.

The structure was solved by direct methods, and calculations were performed on PDP 11/44 and IBM 486 computers.

REFERENCES

[1] G. Levitt: in D. R. Baker, J. G. Fenyes, W. K. Moberg (eds): *Synthesis and Chemistry of Agrochemicals II,* American Chemical Society, Washington, DC, pp. 16–31 (1991).

[2] A. Percival, *Pestic. Sci., 31,* 1991, 569.

[3] Z. M. Li, G. F. Jia, L. X. Wang, C. M. Lai, *Chem. J. Chinese Univ., 15,* 1994, 391.

[4] Z. M. Li, G. F. Jia, L. X. Wang, C. M. Lai, Chinese Patent ZL 93 1 01976.1, 1995.

[5] A. A. Munshi et al., *Indian J. Chem., 1,* 1963, 311.
[6] M. P. Sharma et al., *Indian J. Pharm. Sci., 43,* 1981, 177.
[7] K. A. Jansen, *Acta Chem. Scand., 15,* 1961, 1787.
[8] J. S. Upadhyovy, *J. Chromatogr., 152,* 1987, 595.
[9] A. K. Dimri, S. S. Parmar, *J. Heterocyclic Chem., 15,* 1978, 335.
[10] T. Kuragano et al., Japanese Patent 63/91 390, 1988.