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5,7-Diarylspiro[4,5,6,7-tetrahydrobenzo[d][1,2,3]selena/thiadiazole-6,5′-(hexahydropyrimidine)]-2′,4′,6′-triones/-2′-thioxo-4′,6′-diones from Cyclohexanonedicarboxylates: Part V

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ABSTRACT: *The keto and gem-ester functionalities of cyclohexanonedicarboxylates (***1***) offer a facile route for the synthesis of 5,7-diarylspiro[4,5,6,7-tetrahydrobenzo[d][1,2,3]selena/thiadiazole-6,5′-(hexahydropyrimidine)]-2′,4′,6′-triones (***6*** and ***8***) and -2′-thioxo-4′,6′-diones (***7*** and ***9***). The new compounds were characterized by IR, NMR, and CMR spectral data.* © 1999 John Wiley & Sons, Inc. Heteroatom Chem 10: 17–23, 1999

INTRODUCTION

In our earlier communications, we have reported the synthesis of spiro-pyrimidinetriones, pyrazolidinediones, and isoxazolidinediones by exploiting the *gem*-ester group of dimethyl 2,6-diaryl-4-oxocyclohexane-1,1-dicarboxylates and 1,1-dimethoxycarbonyl-2,6-diaryl-4-thia-4,4-dioxides [1]. In our continued efforts to develop some more interesting heterocycles, we thought it fit to synthesize fused spiro-heterocyclic systems by exploiting the α-ketomethylene functionality presented in the latter compound. To accomplish this, 1,2,3-selena/thiadiazole rings have been chosen to build onto spiro-pyrimidinetriones and thioxopyrimidinediones. Sulfur and selenium being isosteric, if one is replaced by the other in any molecule at the same position, it would give an opportunity for the relative comparison of their chemotherapeutic activities. Moreover, reports about selenium-containing heterocycles are relatively few [2]. In view of the foregoing, we report in this communication the syntheses of hitherto unknown fused 1,2,3-selena/thiadiazole spiro-heterocycles.

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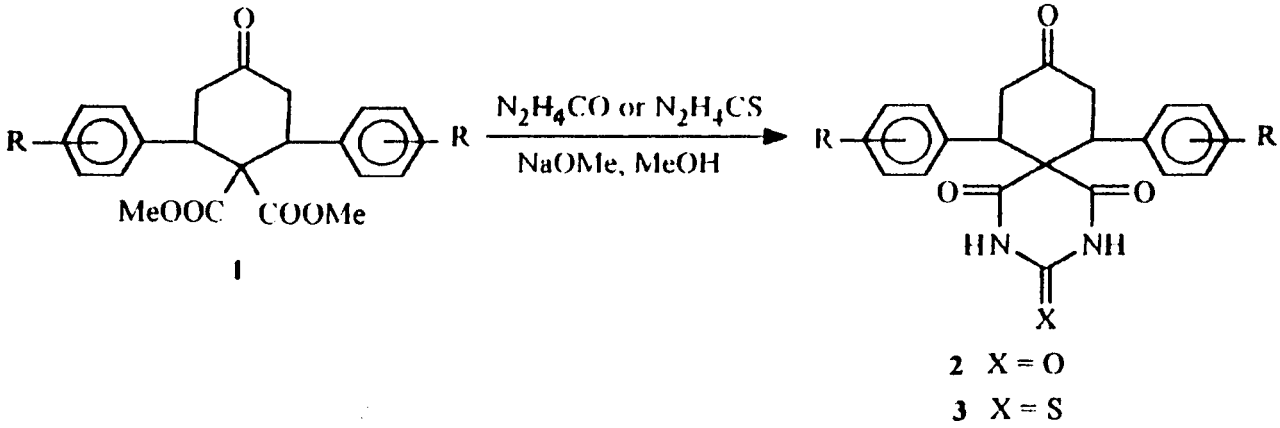
RESULTS AND DISCUSSION

The facile reactivity of active methylene compounds, namely, dimethyl malonate with 1,5-diaryl-1,4-pentadien-3-ones, affords the key intermediates, dimethyl 2,6-diaryl-4-oxocyclohexane-1,1-dicarboxylates (**1**) that were utilized for the synthesis of the target molecules. The condensation of **1** with urea and thiourea gives 7,11-diaryl-2,4-diazaspiro[5,5]-undecane-1,3,5,9-tetraones/-3-thioxo-1,5,9-triones (**2** and **3**) (see Scheme 1).

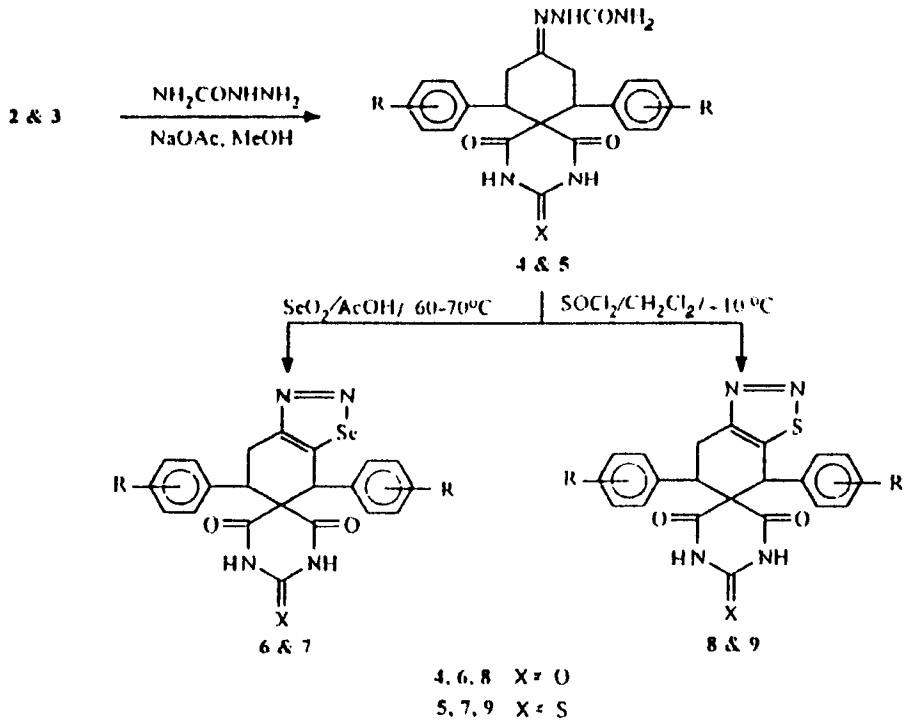
The carbonyl group having an adjacent methylene group in **2** and **3** has facilitated the construction of the selena/thiadiazole ring. The 5,7-diarylspiro[4,5,6,7-tetrahydro[*d*][1,2,3] selena/thiadiazole-6,5′-(hexahydropyrimidine)]-2′,4′,6′-triones (**6** and **8**) have been prepared by the cyclization of the semicarbazones of **2** and **3** (**4** and **5**) with selenium dioxide in acetic acid. On the other hand, the

cyclocondensation of **4** and **5** with thionyl chloride in dichloromethane affords 5,7-diarylspiro[4,5,6,7-tetrahydrobenzo[*d*][1,2,3]selena/thiadiazoles-6,5′-(hexahydropyrimidine)]-2′-thioxo-4′,6′-triones (**7** and **9**) (see Scheme 2). The physical data of these compounds are depicted in Tables 1 and 2.

Thus, the keto and *gem* ester functionalities of cyclohexanone dicarboxylates (**1**) offer a facile route for the synthesis of novel fused spiro-heterocyclic systems. The antimicrobial activity of the compounds **6–9** have been tested for antibacterial activity by the Vincent and Vincent method [3] and antifungal activity by the Horsfall and Rich method [4]. All the tested compounds showed relatively high antibacterial activity against *Staphylococcus aureus* and *Bacillus subtilis* (gram +ve) as against *Escherichia coli* (gram −ve). Compounds having thiophene and chloro and methoxy substituents in the phenyl ring exhibited relatively higher activity than the others. In comparison, 1,2,3-thiadiazoles showed greater activity than the corresponding selenadiazoles. On the other hand, all the compounds **6–9** displayed moderate to high activity against fungi, *Curvularia lunata, Fusarium solani,* and *Helminthos-*



SCHEME 1



SCHEME 2

TABLE 1 Melting Points and Yields of Compounds **4** and **5**

Product No.	*R*	*X*	*Mp (°C)*	*Yield (%)*
4a	H	O	210–212	70
4b	4-OCH_3	O	208–209	61
4c	4-Cl	O	215–217	65
4d	3,4-$(OCH_3)_2$	O	220–221.5	62
4e	4-OCH_3, 3-OCH_2CH_3	O	205–207	64
4f	3,4,5-$(OCH_3)_3$	O	232–234	60
4g	4-$N(CH_3)_2$	O	264–266	66
4h	C_4H_3S	O	176–177.5	71
5a	H	S	271–273	73
5b	4-OCH_3	S	279–281	68
5c	4-Cl	S	280–282	72
5d	3,4-$(OCH_3)_2$	S	276–278	75
5e	4-OCH_3, 3-OCH_2CH_3	S	224–225	70
5f	3,4,5-$(OCH_3)_3$	S	258–260	69
5g	4-$N(CH_3)_2$	S	216–217.5	63
5h	C_4H_3S	S	195–197	66

porium oryzae. However, further studies on bioassay are in progress.

The IR spectra (ν, cm^{-1}) of **4** and **5**, **6** *and* **7**, and **8** and **9** exhibited strong bands in the regions 1690, 3460, and 1420 for CO, NH, and C=S of the pyrimidinetrione / thioxopyrimidinedione moiety. The absence of a band at 1680 for the CO of the cyclohexanone moiety and the presence of medium to strong bands around 3500, 3150, 1715, and 1425 for *NH*CO, CO*NH*$_2$, *CO*NH$_2$, and C=N of the semicarbazone moiety indicates the formation of **4** and **5** [5]. Medium bands observed at 1570 were attributed to the N=N group in **6** and **7**, and **8** and **9**, while the bond at 705 is due to C–S in **8** and **9**. This clearly indicates that semicarbazone and α-methylene groups are involved in the cyclization process.

The ^{1}H NMR spectra (δ) of **4** and **5** may be rationalized assuming the two aryl groups to be diequatorial in confirmity with the *cis*-1,3-arrangement of the substituents as in the cases of **1**, **2**, and **3** [1a,1b]. The pyrimidinetrione / thioxopyrimidinedione moiety of **4** and **5**, which itself is nearly planar, is perpendicular to the average plane of the cyclohexanone moiety (see Figure 1).

The methine (H_A) and methylene (H_M & H_X) protons of **4** and **5** exhibit an AMX splitting pattern. The H_A normally should couple with H_M and H_X and appear as a doublet of doublets at a downfield region. The H_X, due to geminal and vicinal couplings, should also exhibit a doublet of doublets in an upfield position of the spectrum, while the H_M, also for similar reasons, should display a doublet of doublets in between H_A and H_X [1b,6]. In fact, the three doublets of doublets observed in the spectra around 3.60–3.72, 3.12–3.22, and 2.50–2.64 are assigned to H_A, H_M, and H_X, respectively. The coupling constants for them are found to be J_{AM} = 10.50, J_{AX} = 5.25, J_{MX} = 15.50 Hz. Furthermore, three broad singlets observed at 10.80–10.95, 7.70–7.75, and 5.80–5.96 are attributed to CON*H* of the ring and CON*H* and CONH_2 of the semicarbazone moiety that disappeared on deuteration. On the other hand, the PMR spectra of **6** and **7** and **8** and **9** differ very much from that of **4** and **5**. Three different sets of signals are observed for methine and methylene protons. Of the two methine protons, the one adjacent to the double bond C_7-H appears at a downfield region due to the anisotropic effect and showed a singlet at 5.20–5.35. On the other hand, the other methine proton C_5-H adjacent to the methylene group displayed an upfield signal at 3.62–3.85 as a doublet of doublets. The methylene protons at C_4 might have been expected to give two doublets of doublets due to geminal and vicinal couplings. Instead, a multiplet is observed in the region 2.60–2.74. As in the case of **4** and **5**, the two aryl groups in the cyclohexene moiety in its preferred half-chair conformation adopt *cis*-1,3-diequatorial positions, while the pyrimidinetrione/thioxopyrimidinedione moiety, being nearly planar, is almost perpendicular to the average plane of the ring as indicated by the Drieding model (see Figure 2). Furthermore, the 1,2,3-selenadiazole/thiadiazole moiety would also be almost in the same plane as that of the cyclohexene. Thus, it may be observed that the selenadiazole / thiadiazole moiety, which is in the average plane of the cyclohexene half-chair conformation, is perpendicular to the pyrimidinetrione/ thioxopyrimidinedione moiety. The δ_H values observed for **6** and **7**, and **8** and **9** are given in Table 3. The δ_C values for these compounds derived from their ^{13}C NMR spectra have also been incorporated in Table 4.

EXPERIMENTAL

Melting points were determined on a Mel-Temp apparatus and are uncorrected. The purity of the compounds was checked by thin-layer chromatography (silica gel H, BDH, hexane: ethyl acetate, 1:1). The IR spectra were recorded on a Perkin-Elmer Grating Infrared Spectrophotometer, Model 337, in KBr pellets. The ^{1}H NMR spectra were recorded in $CDCl_3$ on a Bruker Spectrospin Varian EM-360 Spectrophotometer with TMS as an internal standard. The elemental analyses were performed by the Regional Sophisticated Instrumentation Centre, CDRI, Lucknow.

The 1,1-dimethyl 2,6-diaryl-4-oxocyclohexane-1,1-dicarboxylates (**1**) were obtained by the double Michael addition of dimethyl malonate to 1,5-diaryl-

TABLE 2 Melting Points, Yields, and Analytical Data of Compounds **6–9**

Product No.	R	X	Mp (°C)	*Yield* (%)	*Mol. Formula* (*Mol. wt*)	*Found* (*Calcd*) (%) *C*	*H*	*N*
6a	H	O	208–211	75	$C_{21}H_{16}N_4O_3Se$ (451.34)	55.65 (55.88)	3.70 (3.57)	12.27 (12.41)
6b	4-OCH_3	O	115–116	70	$C_{23}H_{20}N_4O_5Se$ (511.39)	54.13 (54.01)	3.84 (3.94)	10.83 (10.95)
6c	4-Cl	O	155–157	65	$C_{21}H_{14}Cl_2N_4O_3Se$ (520.23)	48.66 (48.48)	2.86 (2.71)	10.66 (10.76)
6d	3,4-$(OCH_3)_2$	O	160–163	70	$C_{25}H_{24}N_4O_7Se$ (571.45)	52.75 (52.54)	4.30 (4.23)	9.65 (9.80)
6e	4-OCH_3, 3-OCH_2CH_3	O	135–137	67	$C_{27}H_{28}N_4O_7Se$ (599.50)	53.89 (54.09)	4.52 (4.70)	9.47 (9.34)
6f	3,4,5-$(OCH_3)_3$	O	145–147	72	$C_{27}H_{28}N_4O_9Se$ (631.50)	51.20 (51.35)	4.51 (4.46)	8.75 (8.87)
6g	4-$N(CH_3)_2$	O	149–151	68	$C_{25}H_{26}N_6O_3Se$ (537.48)	55.69 (55.86)	4.95 (4.87)	15.81 (15.63)
6h	C_4H_3S	O	88–90	71	$C_{17}H_{12}N_4O_3S_2Se$ (463.39)	44.16 (44.06)	2.49 (2.61)	12.18 (12.09)
7a	H	S	192–193	72	$C_{21}H_{16}N_4O_2SSe$ (467.41)	53.78 (53.93)	3.58 (3.45)	11.81 (11.98)
7b	4-OCH_3	S	180–181	75	$C_{23}H_{20}N_4O_4SSe$ (527.46)	52.13 (52.37)	3.92 (3.82)	10.45 (10.62)
7c	4-Cl	S	179–181	70	$C_{21}H_{14}Cl_2N_4O_2SSe$ (536.30)	47.21 (47.03)	2.48 (2.63)	10.64 (10.44)
7d	3,4-$(OCH_3)_2$	S	168–170	68	$C_{25}H_{24}N_4O_6SSe$ (587.51)	51.30 (51.10)	4.24 (4.11)	9.38 (9.53)
7e	4-OCH_3, 3-OCH_2CH_3	S	187–189	70	$C_{27}H_{28}N_4O_6SSe$ (615.57)	52.46 (52.68)	4.70 (4.58)	9.28 (9.10)
7f	3,4,5-$(OCH_3)_3$	S	158–160	66	$C_{27}H_{28}N_4O_8SSe$ (647.57)	50.24 (50.07)	4.19 (4.35)	8.79 (8.65)
7g	4-$N(CH_3)_2$	S	173–175	65	$C_{25}H_{26}N_6O_2SSe$ (553.54)	54.02 (54.24)	4.85 (4.73)	15.34 (15.18)
7h	C_4H_3S	S	102–103	72	$C_{17}H_{12}N_4O_2S_3Se$ (479.46)	42.40 (42.58)	2.41 (2.52)	11.60 (11.68)
8a	H	O	97–98	55	$C_{21}H_{16}N_4O_3S$ (404.45)	62.16 (62.36)	3.88 (3.98)	13.75 (13.85)
8b	4-OCH_3	O	168–170	60	$C_{23}H_{20}N_4O_5S$ (464.50)	59.61 (59.47)	4.22 (4.33)	12.21 (12.06)
8c	4-Cl	O	138–139	56	$C_{21}H_{14}Cl_2N_4O_3S$ (473.34)	53.06 (53.28)	2.85 (2.98)	12.99 (11.83)
8d	3,4-$(OCH_3)_2$	O	143–145	60	$C_{25}H_{24}N_4O_7S$ (524.55)	57.02 (57.24)	4.47 (4.61)	10.81 (10.68)
8e	4-OCH_3, 3-OCH_2CH_3	O	83–84	63	$C_{27}H_{28}N_4O_7S$ (552.61)	58.83 (58.68)	5.19 (5.10)	10.02 (10.13)
8f	3,4,5-$(OCH_3)_3$	O	105–107	58	$C_{27}H_{28}N_4O_9S$ (584.60)	55.68 (55.47)	4.94 (4.82)	9.47 (9.58)
8g	4-$N(CH_3)_2$	O	147–149	62	$C_{25}H_{26}N_6O_3S$ (490.58)	61.00 (61.20)	5.19 (5.34)	17.28 (17.13)
8h	C_4H_3S	O	138–140	56	$C_{17}H_{12}N_4O_3S_3$ (416.50)	49.20 (49.02)	3.03 (2.90)	13.29 (13.45)
9a	H	S	137–138	68	$C_{21}H_{16}N_4O_2S_2$ (420.51)	59.79 (59.98)	3.95 (3.83)	13.44 (13.32)
9b	4-OCH_3	S	144–146	61	$C_{23}H_{20}N_4O_4S_2$ (480.56)	57.70 (57.48)	4.29 (4.19)	11.55 (11.65)
9c	4-Cl	S	156–158	65	$C_{21}H_{14}Cl_2N_4O_2S_2$ (489.40)	51.70 (51.53)	2.74 (2.88)	11.62 (11.44)

TABLE 2 (*Continued*)

Product No.	R	X	Mp (°C)	*Yield* (%)	*Mol. Formula* (*Mol. wt*)	*Found* (*Calcd*) (%) *C*	*H*	*N*
9d	3,4-$(OCH_3)_2$	S	150–152	65	$C_{25}H_{24}N_4O_6S_2$ (540.62)	55.74 (55.54)	4.34 (4.47)	10.52 (10.36)
9e	4-OCH_3, 3-OCH_2CH_3	S	145–146	66	$C_{27}H_{28}N_4O_6S_2$ (568.67)	57.25 (57.02)	4.83 (4.96)	9.71 (9.85)
9f	3,4,5-$(OCH_3)_3$	S	119–120	64	$C_{27}H_{28}N_4O_8S_2$ (600.67)	53.83 (53.98)	4.82 (4.69)	9.50 (9.32)
9g	4-$N(CH_3)_2$	S	124–125	62	$C_{25}H_{26}N_6O_2S_2$ (506.65)	59.05 (59.26)	5.32 (5.17)	16.43 (16.58)
9h	C_4H_3S	S	127–128	60	$C_{17}H_{12}N_4O_2S_4$ (432.56)	47.42 (47.20)	2.90 (2.79)	12.82 (12.95)

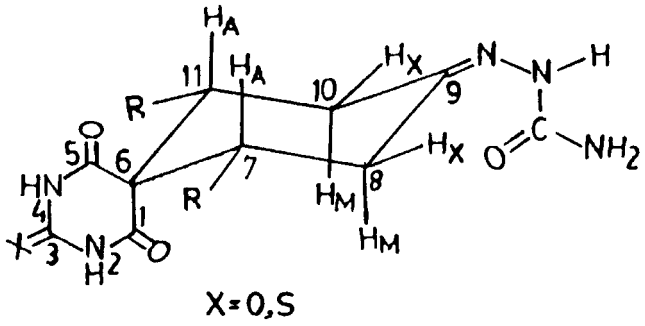


FIGURE 1

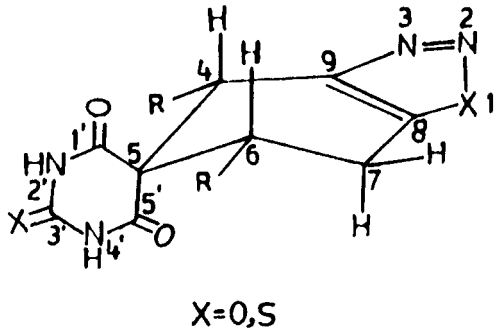


FIGURE 2

1,4-pentadien-3-ones [1a]. The compounds were purified by recrystallization from methanol. **1a**, mp 132–134°C (Ref. [1a], 134–136°C), **1b**, mp 193–195°C (Ref. [1a], 195–196°C), **1c**, mp 195–196°C (Ref. [1a], 197–198°C); **1d**, mp 178–180°C; **1e**, mp 199–201°C; **1f**, mp 148–150°C; **1g**, mp 140–141.5°C; **1h**, mp 148–149°C.

7,11-Diaryl-2,4-diazaspiro[5,5]undecane-1,3,5,9-tetrones/-3-thioxo-1,5,9-triones (**2/3**) were obtained by the cyclocondensation of urea/thiourea to dimethyl 2,6-diaryl-4-oxocyclohexane-1,1-dicarboxylates [1b,7]. The compounds were purified by recrystallization from acetic acid/chloroform. **2a,** mp 290–292°C (Ref. [1b] 292–294°C); **2b**, mp 222–224°C (Ref. [1b] 225–227°C); **2c**, mp 294–296°C (Ref. [1b] 296–298); **2d**, mp 210–212°C; **2e**, mp 216–218°C; **2f**, mp 219–221°C; **2g**, mp 293–295°C; **2h**, mp 210–211°C; **3a**, mp 254–256 (Ref. [7] 250–252°C); **3b**, mp 208–210°C (Ref. [7] 205–207°C); **3c**, mp 194–196°C; **3d,** mp 270–272°C; **3e**, mp 263–265°C; **3f**, mp 225–226°C; **3g**, mp 247–249°C; **3h**, mp 217–219°C.

Semicarbazone of 7,11-Diaryl-2,4-diazaspiro[5,5]undecane-1,3,5,9-tetrones/-3-thia-1,5,9-triones (**4/5**)

A mixture of semicarbazide hydrochloride (15 mmol) and sodium acetate trihydrate (30 mmol) was dissolved in methanol (20 mL), and the residue (NaCl) formed was filtered off. Each compound **2** and **3** (25 mmol) in methanol was added to the filtrate, and the contents were heated on a water bath for 3–5 hours. The reaction mixture was concentrated, cooled, and poured onto crushed ice. The solid obtained was filtered off, dried, and recrystallized from ethanol to obtain pure **4** and **5**.

5,7-Diarylspiro[4,5,6,7-tetrahydrobenzo[d][1,2,3]selenadiazole-6,5'-(hexahydropyrimidine)]-2',4',6'-triones (**6,7**)

The semicarbazone **4** and **5** (10 mmol) was dissolved in glacial acetic acid (20 mL) and warmed gently with stirring to provide a clear solution. Selenium dioxide (10 mmol) in a portionwise manner was then added during a period of 0.5 hour with stirring. The contents were stirred until the evolution of gas ceased, and the deposited selenium was removed by filtration. The filtrate was poured onto crushed ice,

TABLE 3 δ_H Values for **6,7** and **8,9**

Product	*^{1}H NMR ($CDCl_3$)δ*					*Coupling Constants, Hz*		
No.	*NH*	*C_7-H*	*C_5-H_A*	*C_4-H_B*	*C_4-H_X*	*J_{AB}*	*J_{BX}*	*J_{AX}*
6a	11.04	5.53	4.28	3.60	2.78	13.8	14.7	4.5
6b	11.02	5.52	4.08	3.47	2.62	13.8	14.5	4.5
6c	11.01	5.60	4.30	3.48	2.63	13.6	14.5	4.4
6e	11.04	5.58	4.10	3.52	2.67	13.7	14.7	4.5
6g	11.02	5.45	4.32	3.51	2.65	13.6	14.5	4.4
6h	11.02	5.50	4.30	3.57	2.78	13.8	14.5	4.5
7a	11.02	5.45	4.31	3.48	2.65	14.4	15.5	4.2
7c	11.05	5.40	4.35	3.52	2.68	14.3	15.0	4.4
7d	11.00	5.34	4.25	3.47	2.58	14.0	15.4	4.2
7g	10.98	5.51	4.38	3.54	2.59	14.4	15.4	4.3
7h	11.01	5.38	4.29	3.54	2.64	14.3	15.5	4.2
8a	10.8	5.55	4.30	3.55	2.81	12.6	14.1	4.7
8b	11.2	5.42	4.15	3.50	2.74	12.5	14.1	4.7
8c	11.1	5.60	4.19	3.62	2.78	12.3	14.2	4.6
8e	11.0	5.45	4.14	3.53	2.80	12.6	14.2	4.5
8g	10.6	5.44	4.24	3.48	2.82	12.4	14.4	4.6
8h	10.7	5.50	4.32	3.54	2.76	12.6	14.1	4.5
9a	11.4	5.42	4.24	3.60	2.90	11.8	14.5	4.2
9c	10.9	5.48	4.32	3.54	2.95	12.4	14.2	4.0
9d	11.3	5.36	4.18	3.58	2.87	11.9	14.2	4.2
9g	11.5	5.41	4.22	3.49	2.91	12.0	14.4	4.1
9h	11.4	5.42	4.26	3.55	2.85	11.8	14.5	4.0

TABLE 4 δ_C Values for **6,7** and **8,9**

Product	*^{13}C NMR ($CDCl_3$) δ*							
No.	*C_4*	*C_5*	*C_7*	*C_6*	*C_8*	*C_9*	*C_1' & C_5'*	*C_3'*
6a	43.54	57.49	59.77	65.24	129.30	129.19	170.31	149.50
6b	43.52	57.48	59.75	65.22	129.32	129.20	170.29	149.52
6c	43.53	57.46	59.72	65.20	129.30	129.18	170.26	149.54
6e	43.52	57.44	59.76	65.25	129.33	129.22	170.24	149.51
6g	43.50	57.45	59.77	65.26	129.31	129.21	170.22	149.53
6h	43.53	57.42	59.71	65.24	129.30	129.19	170.27	149.50
7a	44.34	59.91	61.23	64.85	129.66	129.41	168.15	148.00
7c	44.32	59.92	61.25	64.84	129.65	129.40	168.17	148.02
7d	44.33	59.90	61.23	64.82	129.63	129.43	168.13	148.05
7g	44.34	59.93	61.25	64.88	129.68	129.46	168.12	148.10
7h	44.33	59.91	61.22	64.86	129.62	129.41	168.14	148.08
8a	43.26	56.46	58.26	61.38	126.62	126.55	174.34	150.20
8b	43.98	55.21	58.64	63.26	127.45	127.30	176.62	149.38
8c	44.05	56.98	59.46	62.66	127.90	127.35	177.36	150.45
8e	43.04	54.49	58.57	62.26	126.96	126.07	175.62	150.34
8g	44.21	56.65	59.76	63.94	127.76	127.15	176.92	150.45
8h	43.60	55.92	58.15	62.61	126.76	126.25	174.92	150.75
9a	44.65	57.45	58.94	63.62	127.77	126.94	175.62	151.12
9c	44.92	56.62	58.06	64.01	127.45	126.38	176.24	152.64
9d	43.74	55.84	57.76	63.98	125.24	126.29	175.38	152.08
9g	44.02	57.49	58.86	63.92	126.69	127.01	175.98	152.96
9h	43.98	56.80	58.11	63.54	125.74	127.15	176.01	151.48

and the collected solid was washed with cold water and sodium bicarbonate solution. The compound thus obtained was purified on a column of silica gel (60–120 mesh, BDH) with hexane:ethyl acetate (1:1) as eluant.

5,7-Diarylspiro[4,5,6,7-tetrahydrobenzo[d][1,2,3]thiadiazole-6,5'-(hexahydropyrimidine)]-2',4',6'-triones (**8,9**)

Each compound **4** and **5** (10 mmol) was added to an excess of thionyl chloride (5 mL) at −10°C in a portionwise manner and allowed to attain room temperature. Then dichloromethane (20 mL) was added, and the resulting mixture was decomposed with saturated sodium carbonate solution. The organic layer was separated and washed thoroughly with water and then dried over anhydrous sodium sulfate. The solvent was evaporated. The crude product obtained was purified by column chromatography using silica gel (60–120 mesh, BDH) with hexane:ethyl acetate (1:1) as eluant.

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