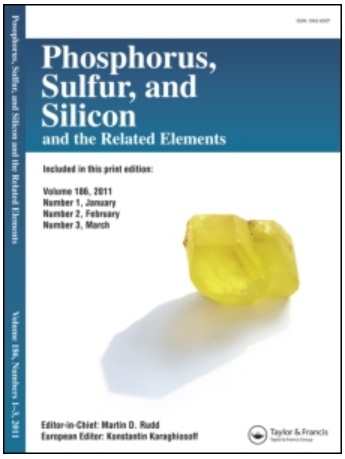

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Synthesis of Novel Isoxazolyl 1,6-Dithia-4,9-diazaspiro[4,4]nonane-3,8-diones and 1-Oxa-6-thia-2,4,9-triazaspiro[4,4]non-2-ene-8-ones

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*The key intermediates, 3-(5-methyl-3-isoxazolyl)-2-arylimino-1,3-thiazolan-4-ones (**3**), were obtained from 3-amino-5-methylisoxazole (**1**) by reaction with chloroacetyl chloride followed by treatment with aryl isothiocyanates. Cyclocondensation of **3** with mercapto acetic acid furnished novel isoxazolyl 1,6-dithia-4,9-diazaspiro[4,4]nonane-3,8-diones (**4**). Cycloaddition of **3** with benzonitrile oxides afforded novel isoxazolyl 1-oxa-6-thia-2,4,9-triazaspiro[4,4]non-2-ene-8-ones (**5**).*

Keywords Cycloaddition; cyclocondensation; isoxazolyl spiro thiazolidinones

INTRODUCTION

The discovery of new agents displaying high antimicrobial activity is an important topic of research in medicinal chemistry.[1] Spirothiazolidinones are heterocyclic nuclei that have stimulated much interest in medicinal and biological chemistry.[2,3] They are known to possess anti-inflammatory,[4] fungistatic,[5] bacteriostatic,[6] anticonvulsant,[7] and antitubercular[8] activities. Similarly, the isoxazole unit constitutes an easily accessible nucleus that is present in a number of natural and pharmacological compounds,[9] displays a wide range of organic reactivities, and could be used as an effective means of preparing new molecular scaffolds.[10] Several of these derivatives are potent antitumor,[11] CNS-active,[12] analgesic,[13] antimicrobial,[14] and chemotherapeutic agents.[15]

Therefore, we thought it useful to construct a system that combines these biolabile rings together in a single molecular framework to see the additive effect towards their biological activities. A literature survey reveals that there is no report on the synthesis of the title molecules so far. In view of these observations and also as a sequel to our search for biologically active nitrogen and sulfur heterocycles[16] linked to an

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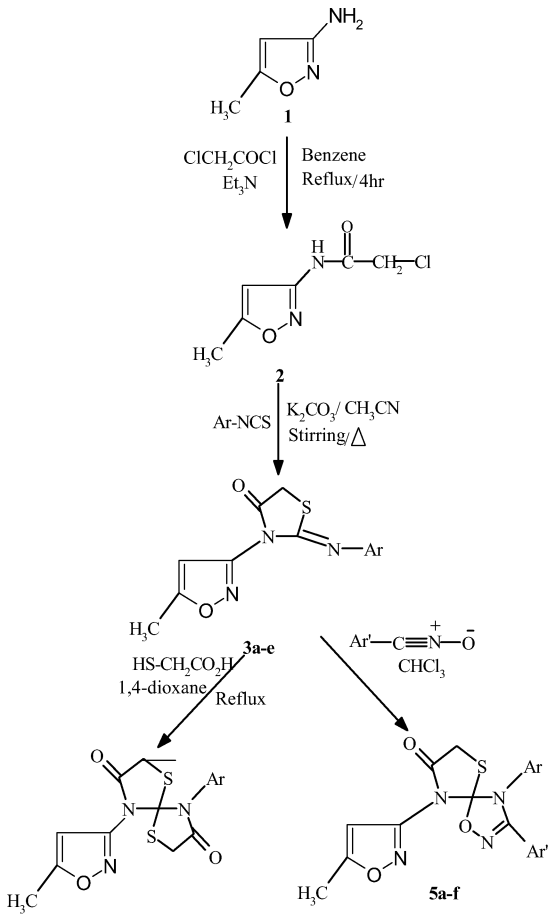
isoxazole ring, we wish to report the synthesis of novel spiro thiazolidinones containing the isoxazole moiety.

RESULTS AND DISCUSSION

The synthetic scheme of compounds **2**, **3**, **4**, and **5** is shown in Scheme 1. The reaction of 3-amino-5-methylisoxazole (**1**) with chloroacetyl chloride in the presence of triethyl amine in dry benzene furnished *N*-1-(5-methyl-3-isoxazolyl)-2-chloroacetamide (**2**). The chloroacetamide (**2**) reacted with aryl isothiocyanates in the presence of K_2CO_3 in CH_3CN to afford the key intermediate viz., 3-(5-methyl-3-isoxazolyl)-2-arylamino-1,3-thiazolan-4-ones (**3**). The mechanism involves the addition of an amide derivative to isothiocyanate in the presence of a base; subsequent cyclization takes place by nucleophilic substation of chlorine by the sulfur atom of isothiocyanate. This mechanism was in agreement with an earlier observation.[17] Cyclocondensation of (**3**) with mercapto acetic acid in 1,4-dioxane led to the formation of novel 4-(5-methyl-3-isoxazolyl)-9-aryl-1,6-dithia-4,9-diazaspiro[4,4]nonane-3,8-diones (**4**). Cycloaddition of (**3**) with benzonitrile oxides, generated in situ, from benzhydroxamoyl chloride in the presence of triethyl amine at ice cold temperature furnished the novel 9-(5-methyl-3-isoxazolyl)-3,4-diaryl-1-oxa-6-thia-2,4,9-triazaspiro[4,4]non-2-ene-8-ones (**5**) (Scheme 1). The structure of the products **2**, **3**, **4**, and **5** have been elucidated on the basis of spectral (IR, ^{1}H NMR, and MS) and microanalytical data (Tables I–V).

Chloroacetamide (**2**) exhibited two strong absorption bands in the IR spectra at 1680 and 3320 cm^{-1} due to C=O and NH functional groups respectively. The ^{1}H NMR spectrum of (**2**) showed two singlets at δ 4.2 and 7.6 due to CH_2 and NH protons, respectively, and the signal at δ 7.6 disappeared upon shaking with D_2O. The mass spectrum of (**2**) displayed the molecular ion $[M+H]^+$ peak at m/z 175. Compounds (**3**) displayed characteristic absorption bands in the IR spectra around 1630 and 1690 cm^{-1} due to C=N and C=O functional groups, respectively, confirming cyclocondensation. The ^{1}H NMR spectra of **3** exhibited a sharp singlet around δ 4.2 due to CH_2 protons. Isoxazole methyl protons exhibited as a sharp singlet at δ2.2, whereas the isoxazole ring proton appeared as a singlet at δ 6.2. Aromatic protons resonated as complex mulitiplet between δ 7.2–7.6. The mass spectrum of the product (**3**) agrees well with the cyclized structure, which showed the molecular ion peak $[M+H]^+$ at m/z 274. The ^{1}H NMR spectra of (**4**) displayed two distinct singlets around δ 4.2 and 4.4 due to methylene protons of thiazolidinone rings, confirming the formation of spirothiazolidinone rings. The mass spectrum of (**4**) confirmed the structure by exhibiting



3 and **4a** Ar, = C_6H_5 **5a** Ar = C_6H_5, Ar' = C_6H_5

3 and **4b** Ar, = 4-$CH_3OC_6H_4$ **5b** Ar = C_6H_5, Ar' = 4-ClC_6H_4

3 and **4c** Ar, = 4-ClC_6H_4 **5c** Ar = 4-$CH_3OC_6H_4$, Ar' = C_6H_5

3 and **4d** Ar, = 4-$CH_3C_6H_4$ **5d** Ar = 4-ClC_6H_4, Ar' = 4-$CH_3OC_6H_4$

3 and **4e** Ar, = 4-BrC_6H_4 **5e** Ar = 4-$CH_3C_6H_4$, Ar' = C_6H_5

5f Ar = C_6H_5, Ar' = 4-$NO_2C_6H_4$

SCHEME 1

the molecular ion peak $[M+H]^+$ at m/z 349. ^1H NMR spectra of (**5**) showed a prominent singlet around δ 4.0–4.3 due to the thiazolidinone ring CH_2 protons. The mass spectrum of (**5**) also agrees well with the structure, which showed the molecular ion peak $[M+H]^+$ at m/z 393, confirming the cycloaddition reaction.

TABLE I Physical Data of Compounds (3a–e)*

Compd.	Mp (°C)	Yield (%)	Mol. Formula (Mol.wt.)	Found (Calcd.) %			
				C	H	N	S
3a	182	85	$C_{13}H_{11}N_3O_2S$ (273)	57.12 (57.14)	4.08 (4.02)	15.44 (15.38)	11.70 (11.72)
3b	132	80	$C_{14}H_{13}N_3O_3S$ (303)	55.49 (55.44)	4.24 (4.29)	13.82 (13.86)	10.51 (10.56)
3c	143	80	$C_{13}H_{10}N_3O_2SCl$ (307)	50.84 (50.81)	3.21 (3.25)	13.72 (13.68)	10.46 (10.42)
3d	168	75	$C_{14}H_{13}N_3O_2S$ (287)	58.56 (58.53)	4.57 (4.52)	14.67 (14.63)	11.19 (11.14)
3e	178	70	$C_{13}H_{10}N_3O_2SBr$ (351)	44.47 (44.44)	2.80 (2.84)	11.92 (11.96)	9.15 (9.11)

*Compounds **3a–e** were recrystallized from aqueous methanol.

TABLE II Physical Data of Compounds (4a–e)*

Compd.	Mp (°C)	Yield (%)	Mol. Formula (Mol.wt.)	Found (Calc.) %			
				C	H	N	S
4a	212	80	$C_{15}H_{14}N_3O_3S_2$ (348)	51.76 (51.72)	4.04 (4.02)	12.09 (12.06)	18.44 (18.39)
4b	160	80	$C_{16}H_{16}N_3O_4S_2$ (378)	50.84 (50.79)	4.28 (4.23)	11.17 (11.11)	16.97 (16.93)
4c	172	75	$C_{15}H_{13}N_3O_3S_2Cl$ (382)	47.16 (47.12)	3.44 (3.40)	10.93 (10.99)	16.71 (16.75)
4d	188	70	$C_{16}H_{16}N_3O_3S_2$ (362)	53.08 (53.03)	4.45 (4.41)	11.66 (11.60)	17.63 (17.67)
4e	195	75	$C_{15}H_{13}N_3O_3S_2Br$ (426)	42.20 (42.25)	3.00 (3.05)	9.88 (9.85)	15.07 (15.02)

*Compounds **4a–e** were recrystallized from aqueous methanol.

TABLE III Physical Data of Compounds (5a–f)*

Compd.	Mp (°C)	Yield (%)	Mol. Formula (Mol.wt.)	Found (Calc.) %			
				C	H	N	S
5a	200	80	$C_{20}H_{16}N_4O_3S$ (392)	61.24 (61.22)	4.02 (4.08)	14.30 (14.28)	8.19 (8.16)
5b	155	75	$C_{20}H_{15}N_4O_3SCl$ (426)	56.37 (56.33)	3.56 (3.52)	13.17 (13.14)	7.54 (7.51)
5c	160	75	$C_{21}H_{18}N_4O_4S$ (422)	59.74 (59.71)	4.28 (4.26)	13.29 (13.27)	7.62 (7.58)
5d	185	70	$C_{21}H_{17}N_4O_4SCl$ (456)	55.22 (55.26)	3.70 (3.72)	12.34 (12.28)	7.04 (7.01)
5e	170	80	$C_{21}H_{18}N_4O_3S$ (406)	62.09 (62.06)	4.44 (4.43)	13.76 (13.79)	7.85 (7.88)
5f	190	70	$C_{20}H_{15}N_5O_5S$ (437)	54.97 (54.91)	3.49 (3.43)	16.04 (16.01)	7.35 (7.32)

*Compounds **5a–f** were recrystallized from benzene.

TABLE IV Spectral Data of Compounds 3 and 4

Compd.	IR (ν_{max} cm^{-1})	^1H NMR (δ ppm)	Mass spectra m/z $[M+H]^+$
3a	1630 C=N 1690 C=O	2.21 (s, 3H, CH_3), 4.21 (s, 2H, CH_2), 6.20 (s, 1H, isoxazole-H), 7.20–7.63 (m, 5H, Ar-H).	262
3b	1635 C=N 1685 C=O	2.31 (s, 3H, CH_3), 3.72 (s, 3H, OCH_3), 4.16 (s, 2H, CH_2), 6.24 (s, 1H, isoxazole-H), 7.00–7.65 (m, 4H, Ar-H).	292
3c	1630 C=N 1695 C=O	2.13 (s, 3H, CH_3), 4.00 (s, 2H, CH_2), 6.00 (s, 1H, isoxazole-H), 7.15–8.00 (m, 4H, Ar-H).	296
3d	1640 C=N 1685 C=O	2.20 (s, 3H, CH_3), 2.40 (s, 3H, CH_3), 4.20 (s, 2H, CH_2), 6.30 (s, 1H, isoxazole-H), 7.00–8.20 (m, 4H, Ar-H).	276
3e	1630 C=N 1680 C=O	2.20 (s, 3H, CH_3), 4.05 (s, 2H, CH_2), 6.40 (s, 1H, isoxazole-H), 7.00–7.55 (m, 4H, Ar-H).	310
4a	1710 C=O 1275 C− S	2.22 (s, 3H, CH_3), 4.21 (s, 2H, CH_2), 4.40 (s, 2H, CH_2), 6.10 (s, 1H, isoxazole-H), 7.21–8.20 (m, 5H, Ar-H).	315
4b	1720 C=O 1280 C− S	2.10 (s, 3H, CH_3), 3.72 (s, 3H, OCH_3), 4.25 (s, 2H, CH_2), 4.45 (s, 2H, CH_2), 6.08 (s, 1H, isoxazole-H), 7.50–8.40 (m, 4H, Ar-H).	345
4c	1715 C=O 1270 C− S	2.20 (s, 3H, CH_3), 4.20 (s, 2H, CH_2), 4.42 (s, 2H, CH_2), 6.12 (s, 1H, isoxazole-H), 7.22–8.00 (m, 4H, Ar-H).	349
4d	1710 C=O 1285 C− S	2.15 (s, 3H, CH_3), 2.32 (s, 3H, CH_3), 4.20 (s, 2H, CH_2), 4.42 (s, 2H, CH_2), 6.15 (s, 1H, isoxazole-H), 7.20–8.00 (m, 4H, Ar-H).	329
4e	1720 C=O 1275 C− S	2.20 (s, 3H, CH_3), 4.25 (s, 2H, CH_2), 4.40 (s, 2H, CH_2), 6.10 (s, 1H, isoxazole-H), 7.50–8.50 (m, 4H, Ar-H).	392

In summary, we have synthesized novel spiro thiazolodines carrying isoxazole moiety by cyclocondensation and cycloaddition reactions. In view of the potential activity of spiro thiazolidinones, we predict that the newly synthesized novel isoxazolyl spiro thiazolidinones may act as drug candidates, and the activity data will be published elsewhere.

EXPERIMENTAL

All the melting points were determined on a Cintex melting point apparatus and are uncorrected. Analytical TLC was performed on Merck precoated 60 F_{254} silica gel plates. Visualization was done by exposing them to iodine vapor. IR spectra (KBr pellet) were recorded on a Perkin-Elmer BX series FT-IR spectrophotometer. ^1H NMR spectra

TABLE V Spectral Data of Compounds 5

Compd.	IR (ν_{max} cm^{-1})	^1H NMR (δ ppm)	Mass spectra m/z $[M+H]^+$
5a	1635 C=N 1680 C=O	2.20 (s, 3H, CH_3), 4.02 (s, 2H, CH_2), 6.10 (s, 1H, isoxazole-H), 7.20–8.20 (m, 10H, Ar-H).	393
5b	1640 C=N 1670 C=O	2.10 (s, 3H, CH_3), 4.18 (s, 2H, CH_2), 6.00 (s, 1H, isoxazole-H), 7.00–8.02 (m, 9H, Ar-H).	427
5c	1640 C=N 1680 C=O	2.32 (s, 3H, CH_3), 3.81 (s, 2H, OCH_3), 4.21 (s, 2H, CH_2), 6.51 (s, 1H, isoxazole-H), 7.50–8.50 (m, 9H, Ar-H).	423
5d	1635 C=N 1685 C=O	2.22 (s, 3H, CH_3), 3.12 (s, 3H, OCH_3), 4.32 (s, 2H, CH_2), 6.20 (s, 1H, isoxazole-H), 7.20–8.50 (m, 8H, Ar-H).	457
5e	1635 C=N 1670 C=O	2.10 (s, 3H, CH_3), 2.50 (s, 3H, CH_3), 4.02 (s, 2H, CH_2), 6.41 (s, 1H, isoxazole-H), 7.10–7.62 (m, 9H, Ar-H).	407
5f	1635 C=N 1695 C=O	2.20 (s, 3H, CH_3), 4.12 (s, 2H, CH_2), 6.00 (s, 1H, isoxazole-H), 7.80–8.62 (m, 9H, Ar-H).	438

were recorded on a Varian Gemini 300 MHz spectrometer. Chemical shift values are given in ppm (δ) with tetramethylsilane as internal standard. Mass spectral measurements were carried out by EI method on a Jeol JMC-300 spectrometer at 70 eV. Elemental analyses were performed on Carlo Erba 106 and Perkin-Elmer model 240 analyzers.

*N*1-[5-Methyl-3-isoxazolyl]-2-chloroacetamide (2)

3-Amino-5-methylisoxazole (**1**) (0.01 mol), chloroacetyl chloride (0.01 mol), and triethyl amine (0.5 mL) were taken in dry benzene (15 mL), and the contents were refluxed with stirring for 4–6 h. The precipitated triethyl amine hydrochloride was removed by filtration. The gummy product obtained after the removal of solvent at ambient temperature was triturated with methanol. Recrystallization of the product was effected from aqueous methanol.

Compound **2**: mp 62°C; Yield: 85%, IR (KBr): 3320 (NH), 1680 (C=O); ^1H NMR (200 MHz, $CDCl_3$): δ 2.21 (s, 3H, CH_3), 4.20 (s, 2H, CH_2), 6.40 (s, 1H, isoxazole-H), 7.60 (s, 1H, NH); MS $[M+H]^+$ m/z: 175; Anal. $C_6H_7N_2O_2Cl$: Found C, 41.40; H, 4.06; N, 16.03. Calcd. C, 41.37; H, 4.02; N, 16.09%.

3- (5-Methyl-3-isoxazolyl)-2-arylimino-1,3-thiazolan-4-ones (3)

To a solution of N1-(5-methyl-3-isoxazolyl)-2-chloroacetamide (**2**) (0.01 mol) in aceto nitrile (15 mL), aryl isothiocyanate (0.01 mol) was added in the presence of K_2CO_3 (0.5 g). The reaction mixture was refluxed while stirring for about 8 h. The solvent was removed under reduced pressure, and the residue was purified by recrystallization from methanol to give (**3a–e**) (Table I).

4-(5-Methyl-3-isoxazolyl)-9-aryl-1,6-dithia-4,9-diazaspiro[4,4]nonane-3,8-dione (4)

A mixture of compound (**3**) (0.01 mol) and mercapto acetic acid (0.01 mol) was dissolved in 1,4-dioxane (15 mL), and the contents were refluxed for 5–8 h. After completion of the reaction (monitored by TLC), the reaction mixture was allowed to cool and was poured over crushed ice. The organic layer was extracted with ethyl acetate (20 mL), washed with 10% sodium bicarbonate solution (1 × 20 mL), and dried over anhydrous sodium sulfate. The solvent was removed under vacuum, and the residue was recrystallized from methanol to give (**4a–e**) (Table II).

9-(5-Methyl-3-isoxazolyl)-3,4-diaryl-(1-oxa-6-thia)2,4,9-triazaspiro[4,4]non-2-ene-8-ones (5)

A mixture of compound **3** (0.01 mol) in dry chloroform (50 mL) was cooled in an ice/salt bath, and a solution of benzhydroxamoyl chloride (0.01 mol) in chloroform (10 mL) was added. Triethyl amine (0.01 mol) in chloroform (10 mL) was added to the reaction mixture at 0°C over a period of 15 min with constant stirring. After the addition was completed, the stirring was continued for another 4 h at 0°C. The chloroform layer was washed with water (2 × 25 mL) to remove triethyl amine hydrochloride, and the organic layer was dried (Na_2SO_4). The solvent was removed at ambient temperature, and the crude product was triturated with light petrol repeatedly to obtain a residue. Recrystallization from benzene gave (**5a–f**) (Table III).

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