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Fluorine containing heterocyclic compounds: synthesis of 6-substituted-2-substituted-aryl-1,2,4-triazolo[5,1-b] 1,3,5-thiadiazin-7-one derivatives

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Abstract

A group of heterocylic compounds of condensed triazole-thiadiazinone derivatives containing fluorine (**IIIa–h**) were obtained in good yields by the reactions of the corresponding 3-aryl-5-mercapto-1,2,4-triazole (**IIa–d**) with *N*-substituted-*N*-chloromethyl carbamoyl chloride (**I**) in the presence of potassium carbonate in *N,N*-dimethylformamide at room temperature. Cyclization of 3-(2,3,5-trifluoro-4-methoxyl)phenyl-5-mercapto-1,2,4-triazole (**IIe**) with **I** was difficult due to an intermolecular hydrogen bond of **IIe**. The structures of the compounds synthesized were confirmed by IR, MS, ^{1}H NMR and elemental analyses. Fungicidal and insecticidal activities of these compounds were also studied.

Keywords: Triazole; Triazole-thiadiazinone; Biological activity

1. Introduction

Triazole derivatives have been reported to have pharmacological, insecticidal, fungicidal and herbicidal activities [1–6,20–22]. Some thiadiazinone derivatives are reported as insect growth regulators [7,8]. In view of these facts and with hope of developing bioactive compounds with high potency, a series of novel heterocyclic compounds containing *s*-triazole and 1,3,5-thiadiazin-4-one nuclei were synthesized and a few of them showed moderate insecticidal, fungicidal and herbicidal activitivies [9]. Organic fluorides have good and extensive biological activities allowing their possible application in pharmaceuticals and pesticides [10]. However, no report describes the effect of fluorine incorporation on the synthesis and bioactivity of previous compounds. It prompted us to synthesize some new fused triazolo-thiadiazinone compounds containing a fluoroaryl moiety, which might be expected to enhance their activities (Scheme 1).

2. Results and discussions

It was easier to prepare the intermediate (**Ia**) by the reaction of *N*-methylformanilide with sulfuryl chloride instead of chlorine [11] (Scheme 2).

The intermediate (**Ib**) was synthesized in 65% yield by the reaction of 1,3,5-triethyl-perhydro-*s*-triazine with bis(trichloromethyl)carbonate instead of phosgene (Scheme 3) [12,23].

The triazoles (**IIa–e**) described in this study were prepared according to the method of Hoggarth [13] and Sandstrom and Wennerberk [14]. Thus, the reaction of aroyl chlorides (**1**) and thiosemicarbazide (**2**) in dried pyridine gave 1-substituted thiosemicarbazides (**3**), which, without purification, were cyclized in refluxing diluted aqueous potassium hydroxide to yield the desired 3-aryl-5-mercapto-1,2,4-triazoles (**IIa–e**) (Scheme 4). The synthesized compounds **IIb–e** were characterized by their IR, ^{1}H NMR, mass spectra and elemental analyses. The IR spectra of compounds **IIb–d** showed NH and SH stretching bands at 3100–3300 and 2600–2700 cm^{-1}, respectively. The ^{1}H NMR spectra of compounds **IIb–d** showed signals at δ 13.7–14.0 ppm attributed to NH and SH of triazole.

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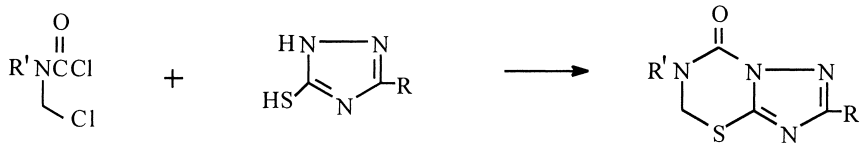
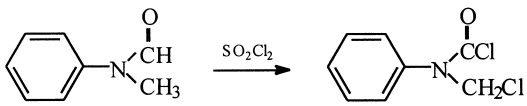


Ia: R'= C_2H_5 **Ib**: R'= C_6H_5 **IIa-d** **IIIa-h**

IIIa R'= C_2H_5 R=2-FC_6H_4, **IIIb** R'= C_2H_5 R=3-FC_6H_4, **IIIc** R'= C_2H_5 R=4-FC_6H_4, **IIId** R'= C_2H_5 R=2,4-diCl-5-F-C_6H_2

IIIe R'= C_6H_5 R=2-FC_6H_4, **IIIf** R'= C_6H_5 R=3-FC_6H_4, **IIIg** R'= C_6H_5 R=4-FC_6H_4, **IIIh** R'= C_6H_5 R=2,4-diCl-5-F-C_6H_2

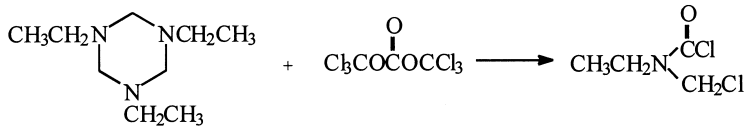
Scheme 1.



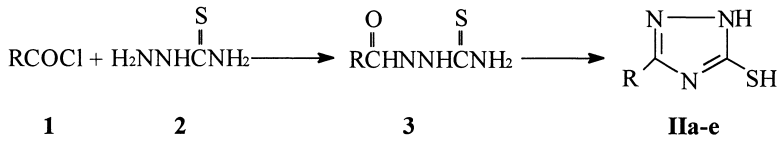
Scheme 2.

1,2,4-Triazolo[5,1-b]1,3,5-thiadiazinone derivatives (**IIIa–h**) were synthesized by the reactions of *N*-chloromethyl carbamoyl chloride (**I**) with 3-fluorophenyl-5-mercapto-1,2,4-triazole in *N*,*N*-dimethylformamide in the presence of potassium carbonate (see Scheme 1). Considering both the difference in reactivity between the chloromethyl and the carbamoyl chloride group [15,24] and in basicity among 1-N, 2-N and 4-N in the *s*-triazole [16,17], we suggest that the cyclization reaction occurs to give expected product **III** as shown in Scheme 5. The ^{1}H NMR spectra of the compounds **IIIa–h** showed a singlet at δ 4.81–5.21 ppm for two protons corresponding to the methylene of thiadiazinone and lacked the NH signal and the SH signal corresponding to a triazole. The IR spectra of compounds **IIIa–h** showed no NH band around 3100–3300 cm^{-1} or SH band around 2600–2700 cm^{-1}, but showed carbamoyl absorption of thiadiazinone around 1715–1735 cm^{-1}. The mass spectrum were consistent with the expected structures (Table 3).

However, a similar reaction with 3-(2,3,5-trifluoro-4-methoxyl)phenyl-5-mercapto-1,2,4-triazole (**IIe**) did not give the expected fused triazolo-thiadiazinone compound. Further, the IR spectra of the product **IIe** showed a broad



Scheme 3.

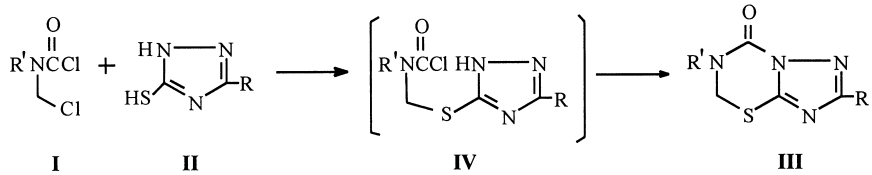


1 **2** **3** **IIa-e**

IIa: R=4-FC_6H_4, **IIb**: R=2-FC_6H_4, **IIc**: R=3-FC_6H_4,

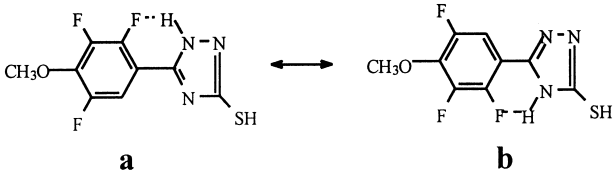
IId: R=2,4-dichloro-5-F-C_6H_2, **IIe**: R=2,3,5-trifluoro-4-CH_3O-C$_6$H

Scheme 4.



I **II** **IV** **III**

Scheme 5.



Scheme 6.

absorption at 2500–3400 cm^{-1} corresponding to a hydrogen bond. The ^1H NMR of compound **IIe** exhibited two broad signals at δ 14.26 and 14.35 ppm for one proton attributed to SH and the chemical shift of corresponding NH has undergone a upfield shift to δ 11.0–11.35 ppm with two singlets (see data given in Section 3). The spectral data of compound **IIe** indicated that two types of hydrogen bond between fluorine in the 2-position of the aromatic ring and hydrogen attached to 2-N or 4-N of the triazole ring were formed in a ratio about 6:4. Many possible three-dimensional molecular conformations and their energies of compound IIe were studied using PCMODEL method (Fourth Edition, June 1990). The calculation results indicated that the structural conformation **a** (E=27.07 kcal/mol) and **b** (E=25.48 kcal/mol) were the most stable with the lowest energies, and conformation **b** most probably the higher concentration of the two isomers. This was consistent with the experiment. Thus, we assume that, due to the strong electron-withdrawing effect of the polyfluorophenyl group, the acidity of NH in the triazole ring would increase and a hydrogen bond is formed between fluorine in the 2-position of aromatic ring and hydrogen attached to 2-N or 4-N of the triazole ring (Scheme 6). This would make cyclization very difficult.

Some of the synthesized **III** type compounds were screened for their antifungal and pesticidal activities. The compounds **IIIe**, **IIIg** and **IIIh** showed weak activities at 50 g/l against *Actinomucor elegans* M-211, *Botrytis cinerea*, *Aspergillus niger*, *Alternaria solani* and *Fusarium oxysporum*, *F. vasinfectum* using the plate technique in an agar medium by measuring the inhibition zone in millimeter [18]. Compound **IIIh** showed 90% insecticidal activity at 300 g/l against house mosquito larvae (*Cluex pipiens molestus* Forskae). None of **IIIa–h** showed activities against *Aphis laburnl* Kaltenbach and *Cirphis unipuncta* Haworth.

2.1. Pests and test methods

2.1.1. Test against Aphis laburnl Kaltenbach

Pea seedlings were immersed in test solution for 20 s, dried and infested with a mixed population. Samples were checked for mortality 24 h after introduction.

2.1.2. Test against Cirphis unipuncta Haworth

Corn leaf-cuts were immersed in test solution for 20 s. Each treated leaf-cut was placed in a Petri dish and two-instar larvae of *C. unipuncta* Haworth were released into each dish. Treated insects were kept at 25°C, 70% relative humidity under lights condition. Mortality was observed 5 days later. Thirty larvae were used for each treatment with two replications.

2.1.3. Test against northern house mosquitoes larvae

A test compound was dissolved in small amount of DMF and diluted with pure water to prepare a test solution of 50 g/l. Thirty third-instar larvae of mosquitoes were released into the test solution (50 ml wide-mouth bottle). Treated insects were kept at 25°C. Mortality was observed 1 day later. Thirty larvae were used for each treatment with two replications.

3. Experimental details

Melting points were taken on a digital melting point apparatus made in Shanghai and were uncorrected. Infrared spectra were measured using a Nicolet 20sx FT-IR instrument. Mass spectra were measured on a Hitachi M80 instrument. ^1H NMR spectra were obtained using a Bruker AC-500 (500 MHz) spectrometer with $CDCl_3$ or DMSO-d_6, using TMS as internal standard. Analysis was undertaken with an Italian MOD 1106 analyzer at the Analysis Center of the East China University of Science and Technology.

3.1. Preparation of N-chloromethyl-N-ethyl-carbamoyl chloride (**Ia**)

To a solution of 39.75 g (0.015 mol) of bis(trichloromethyl)carbonate in 100 ml of chloroform cooled down from −10 to 0°C in a ice-salt bath, 26.65 g (0.015 mol) of 1,3,5-triethyl-hexahydro-*s*-triazine in 20 ml of chloroform was added dropwise below 0°C. The mixture was agitated at room temperature for 1 h, then refluxed for 1 h. Solvent was removed under reduced pressure, and the residue was distilled in vacuo. The product, a colorless liquid, bp: 83–85°C/12 mmHg (bp: 83°C/12 mmHg [12]), was obtained in 65% yield.

Anal. calcd. for $C_4H_7Cl_2NO$: C, 30.80; H, 4.52; N, 8.98%; found: C, 30.92; H, 4.50; N, 8.83%.

3.2. Preparation of N-chloromethyl-N-phenylcarbamoyl chloride (**Ib**)

A solution of 112 g (0.83 mol) of sulfuryl chloride in 70 ml of carbon tetrachloride was added dropwise to a solution of 44.8 g (0.33 mol) *N*-methylformanilide and 4 g of 2,2′-azo-bis-iso-butyronitrile in 230 ml of carbon tetrachloride at 70–75°C over 3–4 h. After addition, the reaction mixture was refluxed for 1–2 h followed by tlc. The reaction was terminated when only trace amount of starting material was left unchanged. The solvent and excess sulfuryl chloride was removed at reduced pressure leaving a syrupy crude material which crystallized on standing. Recrystallization from boiling hexane gave 43 g (64%) of **Ib**; mp: 47.9–50.4°C (mp: 45–46°C [11]).

Anal. calcd. for $C_8H_7Cl_2NO$: C, 46.86; H, 3.44; N, 6.83%; found: C, 46.74; H, 3.48; N, 6.93%.

3.3. General procedures for the preparation of 3-aryl-5-mercapto-1,2,4-triazole (**IIa–e**)

To a stirred, ice-bath solution of the thiosemicarbazide (**2**) (0.02 mol) and pyridine (20 ml) was added dropwise the aroylchloride (**1**) (0.022 mol). The reaction mixture was stirred overnight at room temperature. The excess pyridine was evaporated at reduced pressure, leaving a mixture of the crude aroylthiosemicarbazide (**3**) and pyridine hydrochloride. The mixture was washed with water, removing the pyridine hydrochloride, and the crude aroylthiosemicarbazide (**3**) was collected by filtration. This was heated to reflux in 8% potassium hydroxide or sodium hydroxide solution for approximately 4–5 h. The filtrate was cooled to 0°C. Careful acidification by the dropwise addition of acetic acid afforded the crude solid products, which were recrystallized from ethanol.

3.3.1. 3-(4-Fluoro)phenyl-5-mercapto-1,2,4-triazole (***IIa***)

White powder, 2.88 g (74%), dec.: 272–273°C (dec.: 269–272°C [19]).

3.3.2. 3-(2-Fluoro)phenyl-5-mercapto-1,2,4-triazole (***IIb***)

Colorless plates, 3.15 g (81%), mp: 276–277°C; IR (KBr): 3100, 3050, 2900, 2850, 1620, 1595, 1575, 1495, 1230, 970 cm^{-1}; ^1H NMR (DMSO-d_6): δ 7.35–7.82 (m, 4H), 13.78 (s, 1H), 13.84 (s, 1H); electron impact ms: *m/e* (relative intensity) 195 (100) M^+, 123 (70), 102 (29), 95 (12), 75 (22).

Anal. Calcd. for $C_8H_6FN_3S$: C, 49.22; H, 3.10; N, 21.53%; found: C, 49.02; H, 3.13; N, 21.60%.

3.3.3. 3-(3-Fluoro)phenyl-5-mercapto-1,2,4-triazole (***IIc***)

White powder, 3.00 g (77%), mp: 265–267°C; IR (KBr): 3100, 3050, 2900, 1600, 1570, 1500, 1480, 1240, 980, 960 cm^{-1}; ^1H NMR (DMSO-d_6): δ 7.38 (td, 1H, J=8.5 Hz, J=2.2 Hz), 7.59 (m, 1H), 7.72 (dt, 1H, J=6.1 Hz, J= 2.2 Hz), 7.77 (d, 1H, J=7.9 Hz), 13.76 (s, 1H), 13.81 (s, 1H); electron impact ms: *m/e* (relative intensity) 195 (100) M^+, 136 (41), 122 (36), 95 (12), 95 (23), 75 (15).

Anal. Calcd. for $C_8H_6FN_3S$: C, 49.22; H, 3.10; N, 21.53%; found: C, 49.31; H, 3.12; N, 21.44%.

3.3.4. 3-(2,4-Dichloro-5-fluoro)phenyl-5-mercapto-1,2,4-triazole (***IId***)

White powder, 3.8 g (72%), dec.: 284–285°C; IR (KBr): 3100, 3050, 2850, 2750, 1610, 1590, 1480, 1240, 980, 960 cm^{-1}; ^1H NMR (DMSO-d_6): δ 7.88 (d, 1H, J=9.5 Hz), 8.08 (d, 1H, J=5.8 Hz), 13.88 (s, 1H), 13.95 (s, 1H); electron impact ms: *m/e* (relative intensity) 263 (100) M^+, 265 (68) M^++2, 267 (12) M^++4, 192 (12), 189 (17).

Anal. Calcd. for $C_8H_4Cl_2FN_3S$: C, 36.38; H, 1.53; N, 15.91%; found: C, 36.20; H, 1.52; N, 15.99%.

3.3.5. 3-(2,3,5-Trifluoro-4-methoxyl)phenyl-5-mercapto-1,2,4-triazole (***IIe***)

Yellow needles, 2.97 g (58%), mp: 216–217°C; IR (KBr): 2500–3400, 1630, 1530, 1480, 1360, 1280, 1070, 980, 830 cm^{-1}; ^1H NMR (DMSO-d_6): δ 2.65 (s, 3H), 7.38 (s, 1H), 11.0 (s, NH), 11.35 (s, NH), 14.22 (s, SH), 14.35 (s, SH); electron impact ms: *m/e* (relative intensity) 261 (100) M^+, 188 (11), 174 (36), 125 (11), 99 (14), 88 (36).

Anal. Calcd. for $C_9H_6F_3N_3OS$: C, 41.38; H, 2.32; N, 16.09%; found: C, 41.47; H, 2.30; N, 15.92%.

3.4. General procedures for the preparation of 6-substituted-2-substituted aryl-1,2,4-triazolo-[5,1-b]1,3,5-thiadiazin-7-ones (**IIIa–h**)

A mixture of *N*-substituted-*N*-chloromethyl carbamoyl chloride **I** (0.01 mol), 3-aryl-5-mercapto-1,2,4-triazole **II** (0.01 mol), anhydrous potassium carbonate (0.022 mol) in *N*,*N*-dimethylformamide (10 ml) was stirred at room temperature for 1–2 h, then diluted with water, the resulting precipitate was collected and washed with water. After drying, the crude product was crystallized from ethanol to obtain the desired products **IIIa–h**. Analytical data are recorded in Table 1. The spectroscopic data are given in Tables 2 and 3.

Table 1
Physical data of compounds **IIIa–h**

Compound			mp (°C)	Yield (%)	Formula	Calculated (found (%))		
III	R	R′				C	H	N
a	C_2H_5	2-FC_6H_4	145–146	85	$C_{12}H_{11}FN_4OS$	51.79 (51.57)	3.98 (3.89)	20.13 (20.24)
b	C_2H_5	3-FC_6H_4	160–161	81	$C_{12}H_{11}FN_4OS$	51.79 (51.63)	3.98 (4.06)	20.13 (20.04)
c	C_2H_5	4-FC_6H_4	204–205	86	$C_{12}H_{11}FN_4OS$	51.79 (51.93)	3.98 (3.95)	20.13 (19.99)
d	C_2H_5	2,4-diCl-5-F-C_6H_2	169–171	80	$C_{12}H_9Cl_2FN_4OS$	41.51 (41.64)	2.61 (2.60)	16.14 (16.08)
e	C_6H_5	2-FC_6H_4	218–219	87	$C_{16}H_{11}FN_4OS$	58.89 (58.98)	3.40 (3.37)	17.17 (17.10)
f	C_6H_5	3-FC_6H_4	204–205	83	$C_{16}H_{11}FN_4OS$	58.89 (58.72)	3.40 (3.46)	17.17 (17.31)
g	C_6H_5	4-FC_6H_4	241–242	85	$C_{16}H_{11}FN_4OS$	58.89 (58.71)	3.40 (3.39)	17.17 (17.06)
h	C_6H_5	2,4-diCl-5-F-C_6H_2	197–198	82	$C_{16}H_9Cl_2FN_4OS$	48.65 (48.74)	2.28 (2.30)	14.20 (14.12)

Table 2
^{1}H NMR data of compounds **IIIa–h**

Compound III	^{1}H NMR ($CDCl_3$) δ ppm
a	1.36 (t, 3H, CH_3, J=7.2 Hz), 3.77 (q, 2H, CH_2, J=7.2 Hz), 4.81 (s, 2H, SCH_2), 7.22–8.10 (m, 4H)
b	1.37 (t, 3H, CH_3, J=7.2 Hz), 3.77 (q, 2H, CH_2, J=7.2 Hz), 4.80 (s, 2H, SCH_2), 7.17–7.99 (m, 4H)
c	1.31 (t, 3H, CH_3, J=7.2 Hz), 3.72 (q, 2H, CH_2, J=7.2 Hz), 4.75 (s, 2H, SCH_2), 7.11 (m, 2H), 8.14 (m, 2H)
d	1.33 (t, 3H, CH_3, J=7.2 Hz), 3.73 (q, 2H, CH_2, J=7.2 Hz), 4.78 (s, 2H, SCH_2), 7.54 (d, 1H, J=6.6 Hz), 7.78 (d, 1H, J=9.4 Hz)
e	5.19 (s, 2H, SCH_2), 7.20–7.50 (m, 8H), 8.12 (td, 1H, J=7.5 Hz, J=1.4 Hz)
f	5.21 (s, 2H, SCH_2), 7.20 (m, 1H), 7.41–7.53 (m, 6H), 7.94 (m, 1H) 8.30 (dt, 1H, J=7.7 Hz, J=1.5 Hz)
g	5.18 (s, 2H, SCH_2), 7.14 (t, 2H, J=8.6 Hz), 7.40–7.49 (m, 5H), 8.21 (dd, 2H, J=8.6 Hz, J=5.5 Hz)
h	5.20 (s, 2H, SCH_2), 7.38–7.50 (m, 5H), 7.58 (d, 1H, J=6.7 Hz), 7.83 (d, 1H, J=9.3 Hz)

Table 3
IR and MS spectral data of compounds **IIIa–h**

Compound III	IR (KBr) (cm^{-1})	EI MS *m/e* (%)
a	3020, 2990, 1715, 1615, 1585, 1520, 1500, 1410, 1320, 1190, 1065, 795	278 (86, M^+), 207 (58), 165 (39), 149 (12), 121 (81), 86 (100), 57 (65)
b	2990, 1715, 1615, 1590, 1525, 1500, 1470, 1415, 1310, 1265, 1205, 1180, 875, 790, 740	278 (100, M^+), 207 (46), 165 (43), 149 (26), 121 (73), 86 (47), 57 (49), 56 (14)
c	3010, 2990, 1710, 1605, 1495, 1475, 1415, 1320, 1275, 1215, 845, 745	278 (100, M^+), 207 (48), 165 (43), 149 (28), 121 (82), 86 (40), 57 (42)
d	3010, 2990, 1715, 1600, 1505, 1490, 1470, 1455, 1405, 1335, 1090, 890, 725	350 (5, M^++4), 348 (23, M^++2), 346 (38, M^+), 277 (11), 275 (16), 240 (12), 191 (23), 189 (34), 86 (100), 57 (58), 56 (16)
e	3020, 2950, 1730, 1620, 1580, 1520, 1490, 1440, 1390, 1210, 1060, 770, 760, 740	326 (60, M^+), 207 (48), 165 (4), 149 (10), 121 (19), 105 (100), 104 (40), 86 (40), 77 (17), 57 (45)
f	3020, 1732, 1590, 1520, 1495, 1425, 1350, 1210, 1060, 870, 740	326 (52, M^+), 207 (30), 165 (3), 149 (16), 121 (30), 105 (100), 104 (39), 86 (28), 77 (35), 57 (54)
g	3040, 1730, 1605, 1490, 1430, 1380, 1220, 1070, 850, 750	326 (95,M^+), 205 (39), 165 (5), 149 (26), 121 (38), 105 (100), 104 (39), 86 (20), 77 (43), 57 (57)
h	3050, 1735, 1595, 1495, 1450, 1390, 1315, 1270, 1210, 1090, 1060, 950, 890, 725	395 (68, M^+−1), 397 (44), 276 (11), 241 (13), 191 (10), 189 (15), 105 (100), 104 (46), 86 (27), 57 (29), 50 (14)

Acknowledgements

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