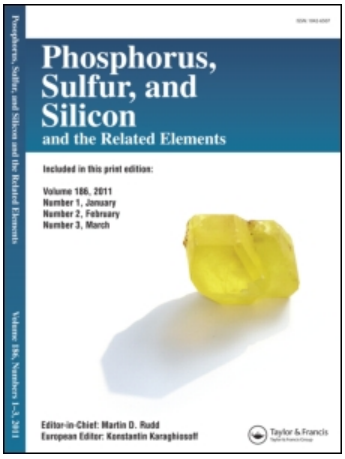

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STUDIES WITH POLYFUCTIONALLY SUBSTITUTED THIAZOLINES AND 1,2,4-TRIAZOLINES: SYNTHESIS AND CHEMICAL REACTIVITY OF 4-ARYLAZO-2 ISOPROPYLOXY-2-THIAZOLIN-5-ONES AND OF 4-ARYLIDENE-2-ISOPROPYLOXY-2-THIAZOLIN-5-ONES

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STUDIES WITH POLYFUCTIONALLY SUBSTITUTED THIAZOLINES AND 1,2,4-TRIAZOLINES: SYNTHESIS AND CHEMICAL REACTIVITY OF 4-ARYLAZO-2 ISOPROPYLOXY-2-THIAZOLIN-5-ONES AND OF 4-ARYLIDENE-2-ISOPROPYLOXY-2-THIAZOLIN-5-ONES

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Ethyl *O*-alkyldithiocarbonates (**1**) reacted with glycine to yield the glycine thiocarboxylates (**3**). On heating in acetic anhydride compounds (**3**) afforded the 2-thiazolin-5-ones (**4**) which could be trapped as 4-arylhydrazones (**4**) and 4-arylidene derivatives (**6**). Compounds (**5a,b**) produced 5-oxo-1,2,4-triazoline-3-carboxylic acid derivatives (**12**) on treatment with amines. The arylidene derivative (**6b**) reacted with amines to yield the imidazolidines (**17**).

Keywords: Thiazolines; azolyl-1,2,4-triazolines; imidazolidines and thione-thiol rearrangements

INTRODUCTION

Polyfunctionally substituted heterocycles are biologically interesting molecules and their chemistry has in the past received interest.[1–3] In the last few years we

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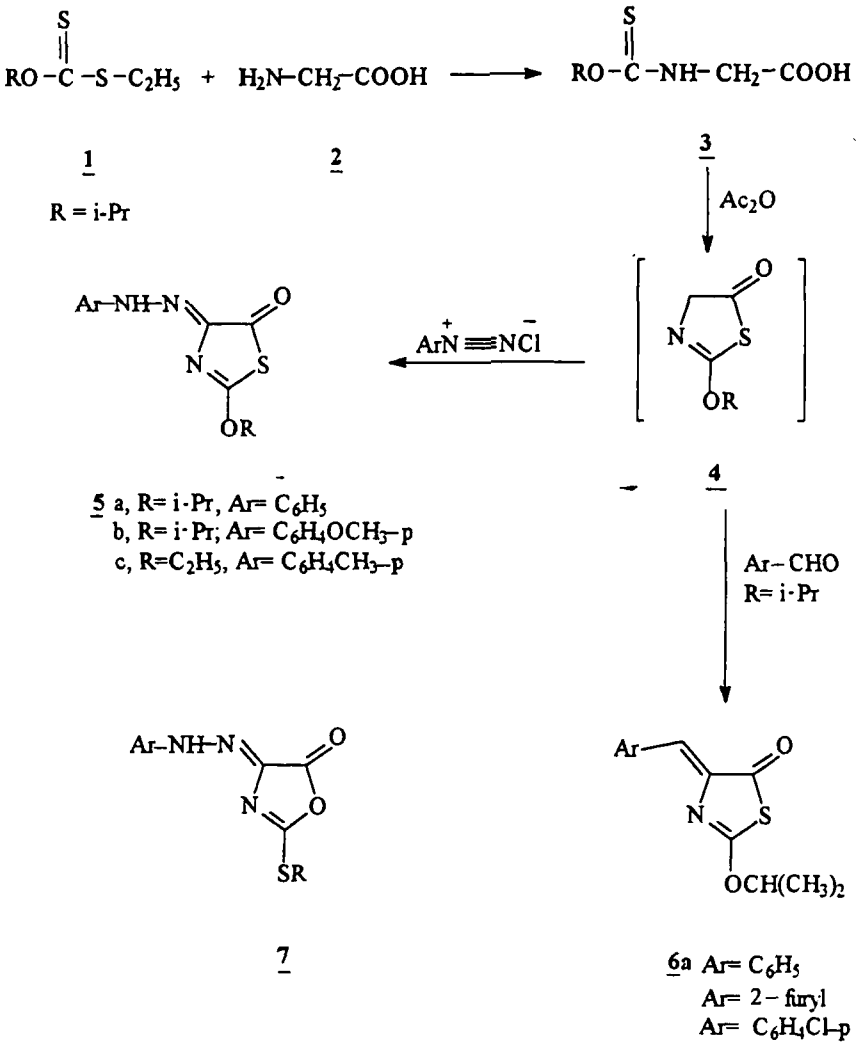
were involved in a program aimed at designing simple and efficient approaches to polyfunctionally substituted heterocycles utilizing inexpensive, readily obtainable starting materials. During this phase of our research a synthesis of thiophenes,[4,5] thiazoles,[6] oxadiazoles,[7] thiadiazoles,[8] pyridazines,[9,10] pyridines[11] and other heterocycles[12–16] were developed. In conjunction with this work we report the synthesis of functionally substituted 2-thiazoline-5-ones and their conversion into several other new functionally substituted azoles.

RESULTS AND DISCUSSION

Ethyl alkyl dithiocarbonates (**1**) have been found to react with glycine (**2**) in refluxing aqueous, potassium hydroxide to yield condensation products that are formulated as **3**. Compound (**3**) readily cyclized on heating with acetic anhydride into the 2-thiazolin-5-one derivatives (**4**). This could never be isolated in pure form but could be trapped as the arylhydrazone (**5a–c**) and the arylidene derivatives (**6a–c**) by coupling with aryldiazonium salts and condensation with aromatic aldehydes respectively. Since alkylthiocarboxylic esters are known to rearrange into alkoxythiones, we have also considered possibility that the reaction product (**7**), formed via such rearrangement. Structure (**5a**) was confirmed based on ^{1}H nmr spectra which revealed the methine proton of the isopropyl group at δ = 5.59 ppm. If the reaction product was **7** this proton should appear at a higher field (ca. 3–4 ppm). Attempted preparation of *N*-(isopropyloxythiocarbonyl)-N-phenylglycine via a sequence similar to that used to prepare **3** failed.

Compounds (**5a,b**) reacted with aromatic amines to yield addition products of aromatic amine and 2-propanethiol elimination. Two structures were considered [*cf.* structures (**11**) and (**12**)]. The imidazolidine structure (**11**) could be readily ruled out based on the preparation of the reaction product from reaction of the known (**13a**)[17] with ethanolic hydrochloric acid. The formation of **12a–C** is assumed to occur via an acyclic intermediate (**8**) which first rearranges into **10** then cyclizes into final product (**12**) via loss of 2-propanethiol. A similar rearrangement reaction has been previously reported on reacting 4-arylhydrazono-2-alkoxy-2-thiazoline with secondary amines[17]. The behavior of **5a,b** toward aromatic amines finds a parallel in the reported reaction of 4-arylhydrazono-2-oxazoline-5-ones[18–20] and of 4-arylhydrazono-2-thiazoline-5-ones with aromatic amines.[15,16,21–23] Similarly, treatment of **5a,b** with aminoheterocyclic compounds yields 1,2,4-triazoline-3-carboxamides (**12d–g**). Although **12a–C** can be obtained from **13a–c** and ethanolic hydrochloric acid the overall yield of their



synthesis via this route is much less than that of their direct formation from **5a,b** and amines. Moreover, compounds **12d–g** could not be obtained via this route as we failed to obtain **13d–g** utilizing a procedure similar to that reported for the synthesis of **13a–c**[15–17]. It has been found that compound (**5c**)[17] on treatment with 5-amino-1,2,4-triazole (**14**) and 5-phenyl-3-aminopyrazole (**15**) gives the 5-ethoxy-1,2,3-triazoles (**16**) and (**17**) respectively. The reaction proceeds via loss of hydrogen sulfide.

Compounds (**5a,b**) reacted with phenols to yield 1,2,4-triazolin-3-carboxylic esters (**18a—d**). Compound (**18a**) could be converted into 1,2,4-triazolin-3-carboxylic acid (**19**). Although compound **19** has been previously reported via rearrangement of 4-phenylazo-2-ethoxy-2-thiazolin-5-one with aqueous sodium hydroxide to our knowledge the formation of **18a—d** via rearrangement of **5a,b** has no similar precedent.

Compound (**6b**) could be also converted into imidazolidines (**21**). Thus, When treated with aniline or 4-phenylthiazole-2-amines, the hydantoin derivatives (**21a,b**) were formed via **20** with loss of 2-propanethiol. Compound **21** also formed in comparable yield via reacting 4-furfurylidene-2-ethoxy-2-thiazolin-5-one with amines.

EXPERIMENTAL

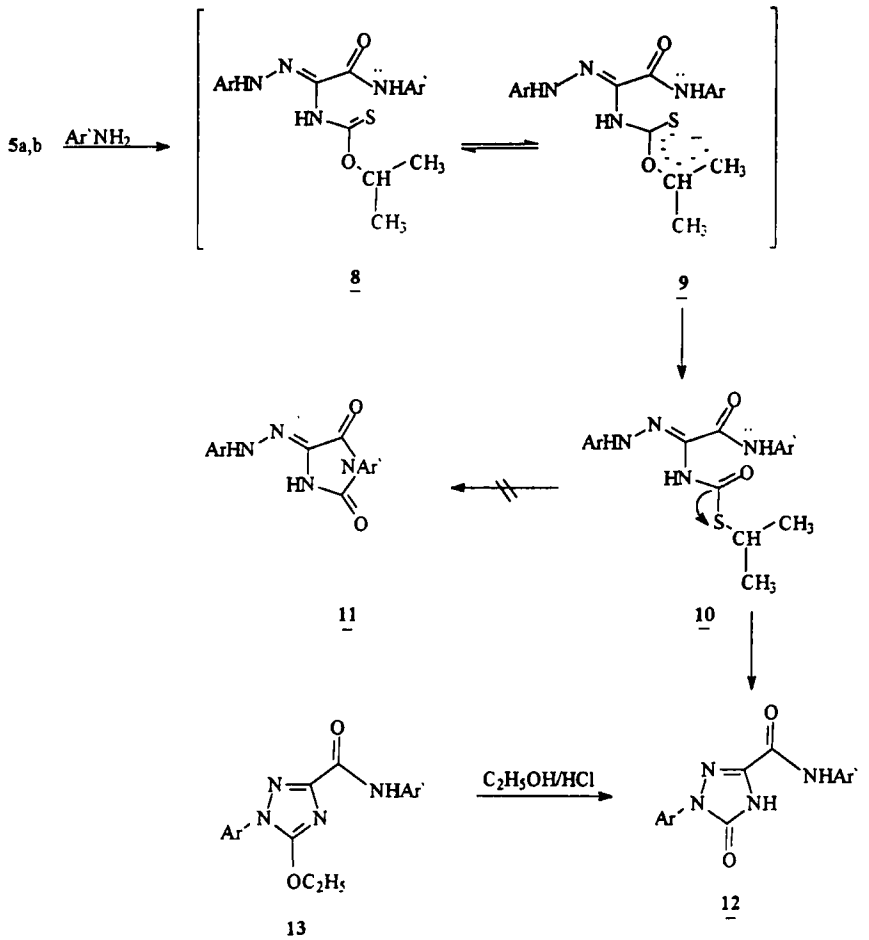
All melting points are uncorrected. Analytical data were obtained from the Microanalytical Data Unit at Cairo University. The IR spectra were obtained on a Pye-Unicam sp-1000 spectrophotometer. ^{1}H nmr spectra were measured in DMSO on a Varian EM-360 MHz, using TMS as internal standard and chemical shifts are expressed as δ ppm. ^{13}C nmr was measured in DMSO on a Bruker AC 250 spectrometer using TMS. Mass spectra were recorded on a Varian MAT 311A spectrometer.

N-(Isopropyloxythiocarbonyl)glycine (*3a*)

A mixture of ethyl *O*-isopropyl dithiocarbonate 1.7 g (0.01 mol) and glycine 0.7 g (0.01 mol) was refluxed in 10% aqueous potassium hydroxide (20ml) for 2h. The reaction mixture was neutralized and the solid was filtered, washed with water, then crystallized from dilute ethanol. **3a**: yield 1 g (56%); mp 125°C; Ir. (KBr) ν 3480–3320 (COOH and NH), 1700(C = O) Cm^{-1}; ^{1}H nmr δ 1.33 (d, 3H, CH_3), 1.35 (d, 3H, CH_3), 4.42 (s, 2H, CH_2), 5.60 (m, 1H, CH), 6.22 (s, 1H, NH), 10.70 (s, 1H, COOH); ms: m/z = 177 (M^+). Anal. Calcd for $C_6H_{11}NO_3S$: C, 40.6; H, 6.2; N, 7.9; S, 18.0 Found: C, 40.7; H, 6.2; N, 7.9; S, 18.2.

4-Arylhydrazono-2-isopropyloxy-2-thiazolin-5-ones (*5a,b*)

Compound (3) 2.2 g (0.01 mol) was heated with 10 ml acetic anhydride at 100°C for 5 min. then treated with the appropriate diazonium chloride [0.01 mol, prepared by adding sodium nitrite 0.5 g (0.01 mol) to the appropriate



a; Ar = Ar` = C_6H_5
b, Ar = $C_6H_4OCH_3$-p ; Ar` = C_6H_5
c, Ar = $C_6H_4OCH_3$-p ; Ar` = $C_6H_4OCH_3$-p
d, Ar = C_6H_5 ; Ar` = 4- phenylthiazol -2-yl
e, Ar = C_6H_5 ; Ar` = 1-phenyl-2,3-dimethylpyrazol-5-one-4-yl
f, Ar = $C_6H_4OCH_3$-p ; Ar` = 4-phenyl-5-methylpyrazol-3-yl
g, Ar = $C_6H_4OCH_3$-p ; Ar`= Pyrid-2-yl

quantity of aromatic amine (0.01 mol) in (5 ml) concentrated hydrochloric acid at 0°C] was added with stirring. After 30 min. the solid product was collected by filtration and crystallized from ethanol.[15]

Compound (**5a**): Yellow crystals; yield 2 g (76%); mp 165°C; Ir. (KBr) ν 3380 (NH), 1710 (C = O) Cm^{-1}; ^1H nmr δ 1.49 (d, 3H, CH_3), 1.51 (d, 3H, CH_3), 5.59 (m, 1H, CH), 7.20–7.89 (m, 5H, Ar-H), 10.30 (br, 1H, NH); ms: m/z = 263 (M^+). Anal. Calcd $C_{12}H_{13}N_3O_2S$: C, 54.8, H, 5.0; N, 16.0; S, 12.2. Found: C, 54.8; H, 5.1; N, 15.8; S, 12.1.

Compound (**5b**): Yellow crystals; yield 1.6 g (57%); mp 153°C; Ir. (KBr): ν 3385 (NH), 1710 (C = O) Cm^{-1}. Anal. Calcd for $C_{13}H_{15}N_3O_3S$: C, 53.2; H, 5.1; N, 14.3; S, 11.0. Found: C, 53.1; H, 5.1; N, 14.3; S, 10.8.

4-Arylidene-2-isopropyloxy-2-thiazolin-5-ones (*6a–c*)

They were prepared by treatment of the appropriate aldehyde (0.01 mol) with **3a** (2.2 g; 0.01 mol) in acetic anhydride (20 ml). The reaction mixture was refluxed for 15 min. After cooling the solid product was collected by filtration and crystallized from dry dioxane.[23]

Compound (**6a**): Colorless crystals; yield 2 g (79%); mp 50°C; Ir. (KBr) ν 1700 (C = O) Cm^{-1}; ^1H nmr δ 1.48 (d, 3H, CH_3), 1.52 (d, 3H, CH_3), 5.65 (m, 1H, CH), 7.00 (s, 1H, Styryl-), 7.45–8.29 (m, 5H, Ar-H). Anal. Calcd for $C_{13}H_{13}NO_2S$: C, 63.2; H, 5.3; N, 5.7; S, 13.0. Found: C, 63.2; H, 5.2; N, 5.8; S, 12.9.

Compound (**6b**): Brown crystals; yield 1.7 g (65%); mp 65°C; Ir. (KBr) ν 1700 (C = O) Cm^{-1}; Anal. Calcd for $C_{11}H_{11}NO_3S$: C, 55.7; H, 4.6; N, 5.9; S, 13.5; Found: C, 55.7; H, 4.3; N, 5.8; S, 13.4.

Compound (**6c**): Colorless crystals; yield 1.8 g (70%); mp 102°C; Ir. (KBr) ν 1695 (C = O) Cm^{-1}; Anal. Calcd for $C_{13}H_{12}NO_2C1S$: C, 55.4; H, 4.3; N, 5.0; S, 11.4. Found: C, 55.4; H, 4.4; N, 5.0; S, 11.2.

1-Aryl-3-(N-substituted Carboxamid)-1,2,4-triazolin-5-ones (*12a–g*)

Method (A): A mixture of **5a,b** (0.01 mol) and the appropriate aromatic or heterocyclic amine (0.01 mol) was heated on a water-bath at 100°C for 2 h. The reaction mixture was triturated with ethanol and the resulting solid was recrystallized from ethanol.

Method (B): 1-Aryl-3-(*N*-arylcarboxamid)-5-ethoxy-1,2,4-triazole (**13**)[15] (0.01 mol) was refluxed in 50 ml of ethanolic hydrochloric acid (20%) for 2h. Ethanol was evaporated and the solid was triturated with water, collected by filtration, then recrystallized from ethanol.

Compound (**12a**): Yield 1.6 g (60%); mp 280°C; Ir. (KBr) ν 3380–3330 (NH), 1690, 1675 (C = O) Cm^{-1}; ^1H nmr δ 7.12–7.51 (m,5H,Ar-H), 7.85–8.19

(m,5H,Ar-H), 10.31 (br,1H,NH), 13.20 (br,1H,NH); ms: m/z = 280 (M^+). Anal. Calcd for $C_{15}H_{12}N_4O_2$: C,64.3; H,4.3; N,20.0. Found: C,64.3; H,4.2; N,19.8.

Compound (**12b**): Yield 2 g (65%); mp 283°C; Ir. (KBr) ν 3390–3330 (NH), 1680, 1665 (C = O) Cm^{-1}. Anal. Calcd for $C_{16}H_{14}N_4O_3$: C,61.9; H,4.5; N,18.1. Found: C,61.8; H,4.4; N, 18.0.

Compound (**12c**): Yield 2.2 g (65%); mp 262°C; Ir. (KBr) ν 3385–3330 (NH), 1700,1670 (C = O) Cm^{-1}. Anal. Calcd for $C_{17}H_{16}N_4O_4$: C, 60.0; H, 4.7; N,16.5. Found: C,59.9; H,4.7; N,16.5.

Compound (**12d**): Yield 1.9 g (55%); mp > 280°C; Ir. (KBr) ν 3380–3330 (NH), 1690, 1670 (C = O) Cm^{-1}; ^1H nmr δ 6.75–7.60 (m,5H,Ar-H), 7.78(s,1H,thiazole-H), 7.92 (m,5H,Ar-H), 10.21 (br,2H,2NH). Anal. Calcd for $C_{18}H_{13}N_5O_2S$: C,59.5; H,3.6; N,19.3; S,8.8. Found: C,59.5; H,3.6; N,19.4; S,8.9.

Compound (**12e**): Yield 1.7 g (45%); mp 270°C; Ir. (KBr) ν 3380–3330 (NH), 1700, 1680 (C = O) Cm^{-1}. Anal. Calcd for $C_{20}H_{18}N_6O_3$: C,61.5; H,4.6; N,21.5. Found: C,61.4; H,4.8; N,21.5.

Compound (**12f**): Yield 1.7 g (46%); mp>280°C; Ir. (KBr) ν 3380–3335 (NH), 1685, 1670 (C = O) Cm^{-1}. Anal. Calcd for $C_{20}H_{18}N_6O_3$: C,61.5; H,4.6; N,21.5. Found: C,61.5; H,4.7; N,21.6.

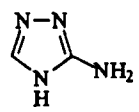
Compound (**12g**): Yield 2 g (63%); mp 217°C; Ir. (KBr) ν 3370–3330 (NH), 1690,1665 (C = O) Cm^{-1}; ^1H nmr δ 3.83 (s,3H,CH_3), 6.95–7.55 (m,4H,pyridine-H), 7.80–8.21 (m,4H,Ar-H),8.20 (br,1H,NH), 10.20 (br,1H,NH); ms: m/z = 311 (M^+); Anal. Calcd for $C_{15}H_{13}N_5O_3$: C,57.9; H,4.2; N,22.5. Found: C,58.2; H,3.8; N,22.4.

1-Aryl-3-(N- substituted carboxamid)-5-ethoxy-1,2,4-triazoles (*16,17*)

A mixture of 4-tolylhydrazono-2-ethoxy-2-thiazolin-5-one (**5c**)[15] 2.8 g (0.01 mol) and the appropriate heterocyclic amine (0.01 mol) was heated on a water-bath at 100°C for 2 h. The reaction mixture was triturated with ethanol and the resulting solid was recrystallized from ethanol as colourless crystals.

Compound (**16**): Yield 1.8 g (60%); mp > 280°C; Ir. (KBr) ν 3380–3350 (NH_2), 1685 (C = O) Cm^{-1}. ^1H nmr δ 1.3 (t,J = 7Hz, 3H, CH_3), 2.28 (s,3H,CH_3), 3.32 (q, J = 7Hz,2H,CH_2), 7.10–8.89 (m,5H,Ar-H), 8.50 (s,1H,NH), 13.00 (br,1H,NH); ms: m/z = 313 (M^+). The ^{13}C nmr spectrum is shown in Scheme 3. Anal. Calcd for $C_{14}H_{15}N_7O_2$: C,53.7; H,4.8; N,31.3. Found: C,53.6; H,4.8; N,31.3.

Compound (**17**): Yield 2.5 g (65%); mp > 280°C; Ir. (KBr) ν 3390–3345 (NH), 1670 (C = O) Cm^{-1}. Anal. Calcd for $C_{21}H_{18}N_6O_2$: C,64.9; H,5.2; N,21.6.



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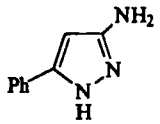

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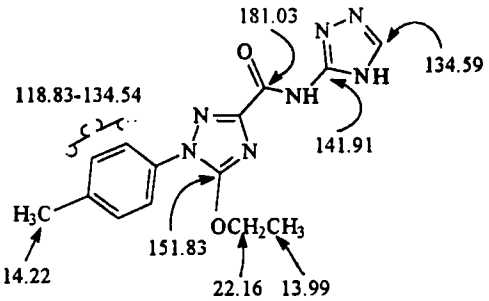

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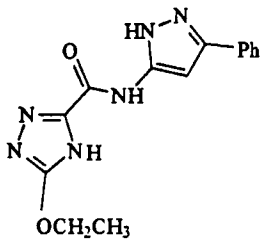

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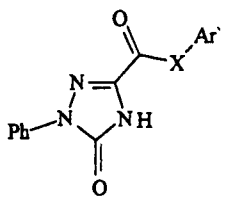
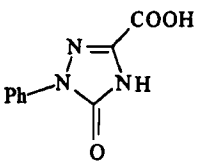
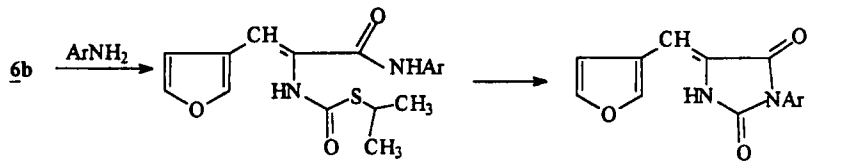




19

18 a, Ar = C_6H_5 ; X= O ; Ar`= 2-naphthyl
b, Ar = C_6H_5 ; X = S ; Ar`= 2-naphthyl
c, Ar = $C_6H_4OCH_3$-p ; X = O; Ar`= C_6H_5
d, Ar = $C_6H_4OCH_3$-p ; X = O; Ar` = C_6H_4Br-p



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21 a, Ar = C_6H_5
b, Ar = 4-phenylthiazol-2-yl

Found C,64.9; H,5.2; N,22.0. *5-Oxo-1-phenyl-1,2,4-triazole-3-carboxylic acid ester* (**18a–d**): A mixture of **5a,b** (0.01 mol) and the appropriate phenol (0.01 mol) was heated on water-bath at 100°C for 2h. The reaction mixture was triturated with ethanol and the resulting solid was crystallized from ethanol. All compounds are colorless.

Compound (**18a**): Yield 1.4 g (45%); mp 240°C; Ir. (KBr) ν 3350 (NH), 1785 (C = O ester; 1710 C = O) Cm^{-1}; ^1H nmr δ 7.00–8.29 (m, 12H,Ar-H), 10.75 (s,1H,NH). Anal. Calcd for $C_{19}H_{13}N_3O_3$: C,68.9; H,3.9; N,12.7. Found: C,68.9; H,3.8, N,12.6.

Compound (**18b**): Yield 1.7 g (51%); mp 263°C; Ir. (KBr) ν 3400(NH), 1795 (C = O thioester), 1710 (C = O) Cm^{-1}; ms: m/z 347 (M^+); Anal. Calcd $C_{19}H_{13}N_3O_2S$: C,65.7; H,3.7; N,12.1; S,9.2. Found: C,65.7; H,3.8; N,12.2; S,9.4.

Compound (**18c**): Yield 1.2 g (42%); mp 213°C; Ir. (KBr) ν 3370 (NH), 1785,1705 (C═O) Cm^{-1}; ^1H nmr δ 3.06 (3H,OCH_3), 6.92–7.59 (m,9H,Ar-H), 10.75(s,1H,NH); Anal. Calcd for $C_{16}H_{13}N_3O_4$: C,61.7; H,4.2; N,13.5. Found: C,61.8; H,4.2; N,13.4.

Compound (**18d**): Yield 1.6 g (42%); mp 139°C; Ir. (KBr) ν 3330 (NH), 1725,1685 (C═O) Cm^{-1}; Anal. Calcd for $C_{16}H_{12}BrN_3O_4$: C,49.2; H,3.1; N,10.8 Found: C,49.3; H,3.2; N,10.9.

5-Oxo-1-phenyl-1,2,4-triazole-3-carboxylic acid (*19*)

A suspension of **18a** 3.3 g (0.01 mol) in 10% ethanolic potassium hydroxide (50 ml) was refluxed for 3h. The resulting reaction mixture was acidified with hydrochloric acid. The resulting solid product was crystallized from benzene as colorless crystals. Yield 0.5 g (25%); mp 152°C; Ir. (KBr) υ 3600-3450 (COOH), 3330 (NH), 1680 (C═O) Cm^{-1}; ^1H nmr δ 7.22–7.65 (m,5H,Ar-H), 9.55 (s,1H,COOH), 10.75 (s, 1H,NH). Anal. Calcd for $C_9H_7N_3O_3$: C,52.7; H,3.4; N,20.5. Found: C,52.7; H,3.4; N,20.6.

1-Aryl-4-furfurylidene-2-hydantion (*21a,b*)

A mixture of **6b** 2.3 g (0.01 mol) and the appropriate amine (0.01 mol) was heated on an oil bath (120°C) for 1h. The reaction mixture was diluted with water, acidified with hydrochloric acid. The solid products were crystallized from ethanol as colorless crystals.

Compound (**21a**): Yield 0.7 g (30%); mp > 280°C; Ir. (KBr) υ 3300 (NH)), 1705, 1675 (C=O) Cm^{-1}; Anal. Calcd for $C_{14}H_{10}N_2O_3$: C,66.1; H,3.9; N,11.0. Found: C,66.2; H,3.7; N,11.3.

Compound (**21b**): Yield 1.1 g (35%); mp > 280°C; Ir. (KBr) υ 3350 (NH), 1710, 1680 (C=O) Cm^{-1}; ^1H nmr δ 6.61–7.19 (m,4H,styryl, Furyl-H), 7.28–7.60 (m,6H,Ar, thiazole-H), 11.19 (br,1H,NH). Anal. Calcd for $C_{17}H_{11}N_3O_3S$: C, 60.5; H,3.3; N,12.5; S,9.5. Found: C,60.4; H,3.2; N,12.5; S,9.5.

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