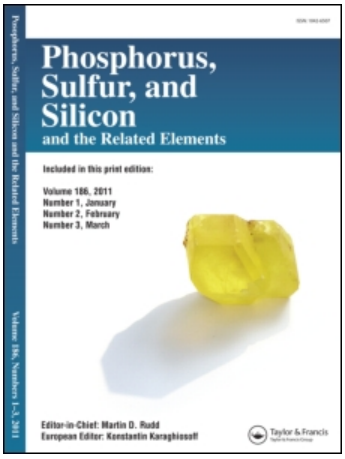

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Synthesis and Studies of Triazolothiadiazines. An Approach Toward New Biologically Active Heterocyclic Compounds

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Synthesis and Studies of Triazolothiadiazines. An Approach Toward New Biologically Active Heterocyclic Compounds

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The first synthesis of the 1,2,4-triazolo[5,1-b][1,3,5] thiadiazine and some of its analogues, possessing a thiadiazino-s-triazole bicyclic ring system was performed starting from 2-benzyltriazole-5-thiol. The title compounds ***6a–g*** *were synthesized via double Mannich reaction in one step in high yields. The route employed the nucleophilic addition of two moles of* CH_2O *onto the NH and SH groups of the triazole ring followed by cyclocondensation with elimination of two* H_2O *molecules to form the thiadiazine derivatives. Examination of the biological activity against the selected fungi and bacteria is the main goal of this article. The tested compounds against fungi revealed that the title compounds were active against most strains of fungi, while the tested compounds were inactive against Gram +Ve and Gram –Ve bacteria. All new synthesized compounds were confirmed using the spectral analyses and the direction of the cyclization was confirmed using the molecular mechanics calculations.*

Keywords Bifunctional; biological screening; cyclocondensation; mannich reaction; synthesis; thiadiazines

INTRODUCTION

The first 1,2,4-triazolo[3,4-b]-1,3,4-thiadiazine A system has been synthesized from 4-amino-3-mercapto-1,2,4-triazoles with α-haloketones[1–14] or α-haloacetals.[1–3,8,13] Several methods for the synthesis of triazolothiadiazines have been adopted also using the cyclocondensation of triazole derivatives with variant chemical reagents.[15–18] Recently, 1,2,4-triazolo[3,4-b][1,3,5]thiadiazines have been reported using

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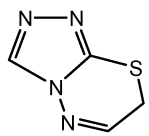
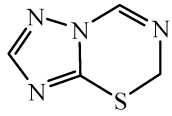


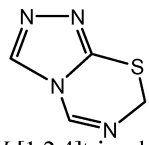
7*H*-[1,2,4]triazolo[3,4-*b*]-[1,3,4]thiadiazine

A



5*H*-[1,2,4]triazolo[5,1-*b*]-[1,3,5]thiadiazine

B



7*H*-[1,2,4]triazolo[3,4-*b*]-[1,3,5]thiadiazine

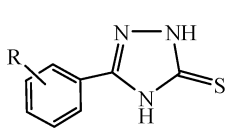
C

CHART 1

Mannich reaction in the presence of acid catalyst.[19–22] To the best of our knowledge, we have found in literature only three classes of the triazolothiadiazine heterocycles. These three classes were categorized as class A, B, and C, Chart 1.

It is well known that Mannich reaction represents a way for introducing a dialkylamino alkyl side chain onto compounds, which contain at least one active hydrogen atom. On the other hand, a double Mannich reaction occurs if the starting compound contains two active adjacent hydrogens.[22,23] Recently, we have reported that 5-phenyl-2H-1,2,4-triazole-3(4H)-thione was reacted with primary amines to give the cyclized triazolo[5,1-b][1,3,5]-thiadiazines via a double Mannich reaction.[23] We have developed an efficient strategy for the synthesis of the triazolothiadiazines from selected triazole compounds. These new heterocyclic systems could be prepared via double Mannich reaction in relatively high yield.[23] Much more work on Mannich cyclization reaction obtained from a variety of di-functional or poly-functional compounds has appeared during the past few years. We have recently reported that these classes of triazolothiadiazines were first synthesized by a modification of Mannich reaction using di-functional compounds, Chart 2.[23]

It has also been reported that the double Mannich reaction of 5-aryl-2H-1,2,4-triazole-3(4H)-thiones with primary aromatic amines in the presence of acid catalyst afforded the 1,2,4-triazolo[3,4-b][1,3,5]thiadiazines (**4a–e**), Scheme 1.[19–22]



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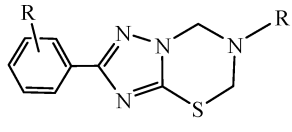

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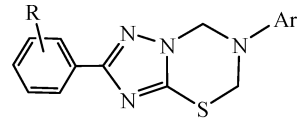

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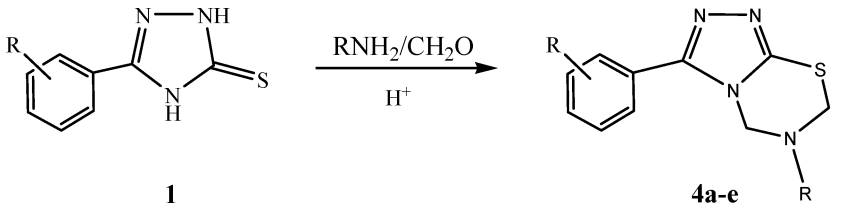
CHART 2

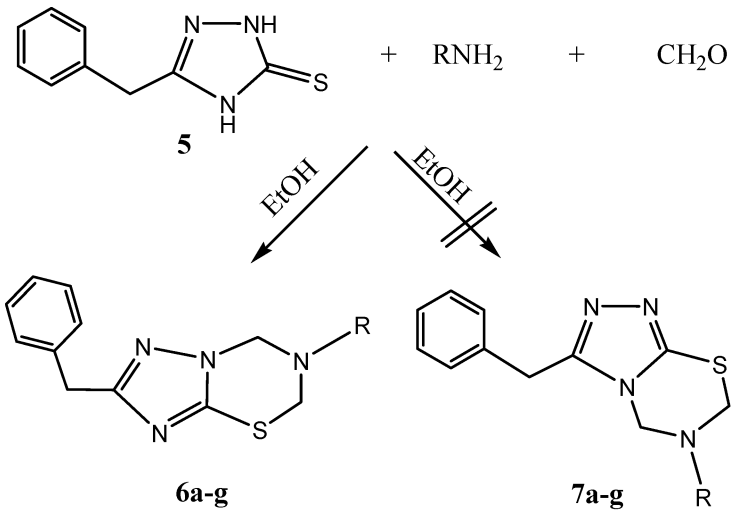


1 **4a-e**

SCHEME 1

RESULTS AND DISCUSSION

Within our continuing research in the field of the synthesis of fused heterocycles using Mannich type reaction we have recently focused our attention on the investigation of the synthesis of novel triazolothiadiazines,[22] triazolothiadiazoles[24] and their analogues as interesting synthetic targets. These results prompted us to study the behavior of 5-benzyl-2H-1,2,4-triazole-3(4H)-thione (**5**) with both primary aliphatic and aromatic amines under Mannich reaction conditions. However, treatment of triazole **5** with formaldehyde and aliphatic amines like methylamine, ethylamine, iso-propylamine, *n*-butylamine and benzylamine or aromatic amine like *p*-toluidine in ethanol gave the corresponding **6a–g** rather than the formation of the isomeric products **7a–g**, Scheme 2.



5

6a-g **7a-g**

6a; R = CH_3, **6b**; R = CH_2CH_3, **6c**; R = $CH(CH_3)_2$, **6d**; R = $(CH_2)_3CH_3$
6e; R = $CH_2CH(CH_3)_2$, **6f**; R = CH_2Ph, **6g**; C_6H_4-CH_3-*p*

SCHEME 2

TABLE I The Molecular Mechanical Calculations of Compounds 6a–g and 7a–g

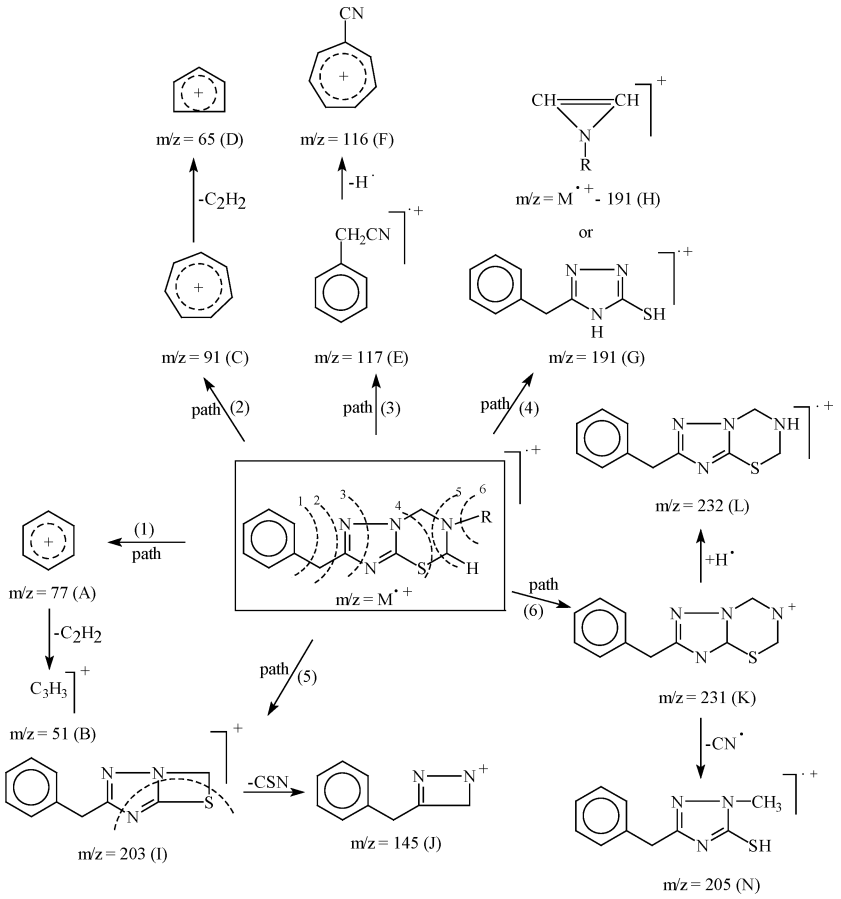
Compound	R	E (k.cal./mol.) of **6a–g**	E (k.cal./mol.) of **7a–g**
6a	CH_3	25.785	26.775
6b	C_2H_5	28.166	29.099
6c	$CH(CH_3)_2$	30.781	31.427
6d	$(CH_2)_3CH_3$	29.898	30.761
6e	$CH_2CH(CH_3)_2$	30.746	30.997
6f	CH_2Ph	28.167	31.615
6g	4-$CH_3C_6H_4$	26.932	27.541

Formation of **6a–g**, rather than **7a–g**, could be attributed to the higher basicity of *N*-1 than *N*-4 of the triazole ring. Besides, the structures of the products **6a–g**, were further attributed to Molecular Mechanical calculations, Table I. While using acid catalyst in the reaction causes the protonation of the *N*-1, which is higher in basicity and leads to the direction of the cyclization, occurs at *N*-4 position.[25] This may be explain the formation of the thiadiazines **7a–g** in the presence of acid catalyst. Details on the cyclization of thiazolotriazoles were summarized in our previous work which supported the formation of thiadiazine derivatives **6a–g** rather the formation of **7a–g** in neutral medium.[26]

The IR spectra of **6a–g** showed the following bands at 3030 (C—H aromatic), 2960 (C—H aliphatic), 1600 (C=N) and at 1580, 1500 cm^{-1} (C=C aromatic skeleton). The ^{1}H-NMR spectra of **6a–g** in $CDCl_3$ showed three singlets signals at δ 5.5–4.9, 4.9–4.6, and δ 4.0 attributed three (CH_2) groups, in addition to the aromatic protons at the expected chemical shift at δ 7.4–6.7 ppm.

The NMR spectral data of compound **6a** (R = CH_3), was compared with the estimated NMR data obtained from ChemDraw Ultra 8.0 and both isomers **6a** or **7a** are similar to the prepared **6a**. These results encouraged use to use the molecular mechanics calculation (MME) to minimize which isomer is most predominant to obtain **6a** or **7a**. From the data obtained in Table I, we can conclude that the isomers **6a–g** were most favorable to be cyclized rather than the isomers **7a–g**.

The fragmentation pattern of the compound **6a** showed the molecular ion peaks at m/z 246 (100%), 260 (100%), 274 (100%), 288 (100%), 288 (100%), 322 (86%), 322 (100 %) respectively, the other fragment peaks (A-N) were in agreement with the proposed fragmentation pattern, Scheme 3.[21]



SCHEME 3

Biological Screening

One of the most interest purposes of the present work was to synthesize new class of heterocyclic thiadiazine compounds, which might be of biological interest. Accordingly, some of these compounds were selected and screened in vitro for their antimicrobial activity against four strains of bacteria [Bacillus cereus, Micrococcus luteus (Gram +ve bacteria) and Escherichia coli, Serratia (Gram –ve bacteria)] and ten strains of fungi (Aspergillus niger, Candida albicans, Chrysosporium tropicum, Fusarium oxysporum, Geotrichum candidum, Microsporum nanum, Trichophyton rubrum), Table II.

TABLE II Antifungal Activity of Compounds 6a–g Expressed in Growth Inhibitory Zone

Fungal species	(a)	(b)	(c)	(d)	(e)	(f))	(g)	(h)	(i)	(J)
Compounds										
Clotrimazol	13	15	10	20	20	15	13	10	11	17
DMSO	0	0	0	0	0	0	0	0	0	0
6a	0	0	0	10	8	10	7	8	0	15
6b	0	0	0	8	8	10	7	7	0	18
6c	0	0	0	10	0	10	7	10	0	15
6d	0	0	12	8	0	10	0	7	0	15
6e	0	0	12	8	0	12	0	12	0	15
6f	0	0	10	8	0	10	0	10	0	12
6g	0	0	10	10	8	10	0	0	0	12

(a) Aspergillus flavus; (b) Aspergillus fumigatus; (c) Aspergillus Niger; (d) Aspergillus var. Albus; (e) Candida albicoms; (f) Chrysosponium tropicum; (g) Fusarium oxysporum; (h) Geotrichum candidum; (i) Microsporum nanum; and (j) Trichophytom rubrum.

The antibacterial and antifungal activities of the tested compounds were evaluated by wells method[27] (5 mm) with 10 microliter/well of (1% concentration) for fungi and 20 microliter/well of (1% concentration) for bacteria using DMSO as a solvent. The inhibition zones (mm) obtained in comparison with those of clotrimazol were summarized in Table II. These results revealed that the most tested compounds were active against most strains of fungi, while all the tested compounds were inactive against Gram +ve and Gram –ve bacteria.

EXPERIMENTAL

Melting points were recorded on a Gallencamp melting point apparatus. Infrared spectra (IR) were measured on a Shimadzu 470 IR spectrometer (KBr, max in cm^{-1}). ^{1}HNMR Spectra were recorded at room temperature on a Varian EM-390, 90 MHz Spectrometer or on a Jeol LA 400 MHz FT-NMR spectrometer. Chemical shifts are denoted in δ units (ppm), relative to tetramethylsilane (TMS) as internal standard, J values are given in Hz. $CDCl_3$ is used as a deuterated solvent unless otherwise stated. MS Spectra was obtained using a JEOL JMS-600 mass spectrometer. Elemental analyses were recorded on a Perkins Elmer 240C elemental analyzer.

Synthesis of 2-Benzyl-6-substituted-6,7-dihydro-5H-[1,2,4]-triazolo[5,1-b][1,3,5]-thiadiazines (6a–g), General Procedure

A mixture of 5-benzyl-2H-1,2,4-triazole-3(4H)-thione (1.91 g, 10 mmols) with formaldehyde (Formalin 40%) (2 ml) and primary amines (10 mmols) was stirred for 5 h at room temperature, then the crude product was crystallized from benzene-cyclohexane to give the targeted compounds **6a–g** as colorless needles in 64–80% yield. The R_f values were measured at ambient temperature using benzene-ethylacetate mixture as an eluent in ratio (6:4).

Synthesis of 2-Benzyl-6-methyl-6,7-dihydro-5H-[1,2,4]triazolo-[5,1-b][1,3,5]-thiadiazines (6a)

R = CH_3. This was obtained as colorless crystals in 70% yield, m.p. 119°C, R_f = 0.4. IR (KBr) ν cm^{-1} = 3030 s (C—H aromatic), 2980 m, 2870 m (C—H aliphatic), 1600 m (C=N), 1560 s, 1510 s, 1460 s, (C=C aromatic skeleton), 1470 m, 1410 m, 1220 m (C—H, C—N), 730 s, 700 s (aromatic). ^{1}H NMR ($CDCl_3$, 90 MHz) δ = 7.4–7.2 (m, 5H, aromatic-H), 4.9 (s, 2H, NCH_2N), 4.6 (s, 2H, SCH_2N), 4.0 (s, 2H, $\mathbf{CH}_2$Ph), 2.6 (s, 3H, CH_3). The MS-EI m/z (%), M^+ = 246 (100%), 116 (58%), 91 (36%), 65 (6%), 203 (17%), 117 (14%), 145 (9%). Elemental analysis for $C_{12}H_{14}N_4S$ (246.34); Calcd: C; 58.51, H; 5.72, N; 22.74, S; 13.01%. Found: C; 58.72, H; 5.55, N; 22.77, S; 12.67%.

Synthesis of 2-Benzyl-6-ethyl-6,7-dihydro-5H-[1,2,4]triazolo-[5,1-b][1,3,5]-thiadiazines (6b)

R = C_2H_5. This was obtained as colorless crystals in 76% yield, m.p. 78°C, R_f = 0.43. IR (KBr) ν cm^{-1} = 3030 s (C—H aromatic), 2980 m, 2870 m (C—H aliphatic), 1600 m (C=N), 1570 s, 1510 s, 1450 s, (C=C aromatic skeleton), 1480 m, 1410 m,1220 m (C—H, C—N), 740 s, 700 s (aromatic). ^{1}H NMR ($CDCl_3$, 400 MHz) δ = 7.3–7.2 (m, 5H, aromatic-H), 5.0 (s, 2H, NCH_2N), 4.6 (s, 2H, SCH_2N), 4.0 (s, 2H,$\mathbf{CH}_2$Ph), 2.8 (s, 2H, CH_2), 1.1 (t, J = 7.0 Hz, 3H, CH_3). The MS-EI m/z (%), M^+ = 260 (100%), 116 (50%), 91 (31%), 65 (4%), 203 (12%), 117 (11%), 145 (9%). Elemental analysis for $C_{13}H_{16}N_4S$ (260.36); Calcd: C; 59.97, H; 6.18, N; 21.52, S; 12.32%. Found: C; 60.51, H; 5.62, N; 21.41, S; 11.98%.

Synthesis of 2-Benzyl-6-isopropyl-6,7-dihydro-5H-[1,2,4]-triazolo[5,1-b][1,3,5]-thiadiazines (6c)

R = $CH(CH_3)_2$. This was obtained as colorless crystals in 64% yield, m.p. 73°C, R_f = 0.45. IR (KBr) ν cm^{-1} = 3030 s (C—H aromatic), 2980 m, 2870 m (C—H aliphatic), 1610 m (C=N), 1580 s, 1520 s, 1460 s, (C=C aromatic skeleton), 1470 m, 1420 m,1220 m (C—H, C—N), 730 s, 700 s

(aromatic). ^{1}H NMR ($CDCl_3$, 90 MHz) $\delta = 7.4$–7.2 (m, 5H, aromatic-H), 5.1 (s, 2H, NCH_2N), 4.7 (s, 2H, SCH_2N), 4.0 (s, 2H,$\mathbf{CH}_2$Ph), 3.2–2.9 (m, 1H, CH), 1.2 (d,$J = 7.0$ Hz, 6H, 2 CH_3). The MS-EI m/z (%), $M^+ = 274$ (100%), 116 (67%), 91 (51%), 65 (6%), 203 (12%), 117 (14%), 145 (12%). Elemental analysis for $C_{14}H_{18}N_4S$ (274.39); Calcd: C; 61.22, H; 6.60, N; 20.42, S; 11.69%. Found: C; 61.72, H; 7.11, N; 20.30, S; 11.24%.

Synthesis of 2-Benzyl-6-butyl-6,7-dihydro-5H-[1,2,4]triazolo-[5,1-b][1,3,5]-thiadiazines (6d)

R = $(CH_2)_3CH_3$. This was obtained as colorless crystals in 65% yield, m.p. 64°C, $R_f = 0.58$. IR (KBr) ν cm^{-1} = 3030 s (C—H aromatic), 2980 m, 2870 m (C—H aliphatic), 1600 m (C=N), 1560 s, 1510 s, 1460 s, (C=C aromatic skeleton), 1480 m, 1410 m,1210 m (C—H, C—N), 740 s, 710 s (aromatic). ^{1}H NMR ($CDCl_3$, 90 MHz) $\delta = 7.4$–7.2 (m, 5H, aromatic-H), 5.0 (s, 2H, NCH_2N), 4.7 (s, 2H, SCH_2N), 4.0 (s, 2H, $\mathbf{CH}_2$Ph), 2.7 (t,$J =$ 7.0 Hz, 2H, CH_2), 1.7–1.2 (m, 4H, 2 CH_2), 1.9 (t,$J = 7.0$ Hz, 3H, CH_3). The MS-EI m/z (%), $M^+ = 288$ (100%), 116 (51%), 91 (51%), 65 (6%), 203 (14%), 117 (14%), 145 (11%). Elemental analysis for $C_{13}H_{16}N_4S$ (288.42); Calcd: C; 62.47, H; 6.99, N; 19.43, S; 11.12%. Found: C; 62.00, H; 6.78, N; 19.16, S; 10.96%.

Synthesis of 2-Benzyl-6-isobutyl-6,7-dihydro-5H-[1,2,4]-triazolo-[5,1-b][1,3,5]-thiadiazines (6e)

R = $CH_2CH(CH_3)_2$. This was obtained as colorless crystals in 73% yield, m.p. 143°C, $R_f = 0.63$. IR (KBr) ν cm^{-1} = 3030 s (C—H aromatic), 2980 m, 2870 m (C—H aliphatic), 1610 m (C=N), 1570 s, 1510 s, 1460 s, (C=C aromatic skeleton), 1480 m, 1410 m,1220 m (C—H, C—N), 730 s, 700 s (aromatic). ^{1}H NMR ($CDCl_3$, 400 MHz) $\delta = 7.3$–7.1 (m, 5H, aromatic-H), 4.9 (s, 2H, NCH_2N), 4.6 (s, 2H, SCH_2N), 4.0 (s, 2H,$\mathbf{CH}_2$Ph), 2.5 (d,$J = 7.0$ Hz, 2H, CH_2), 1.9–1.6 (m, 1H, CH), 0.9 (d,$J = 7.0$ Hz, 6H, 2 CH_3). The MS-EI m/z (%), $M^+ = 288$ (100%), 116 (74%), 91 (76%), 65 (10%), 203 (15%), 117 (17%), 145 (14%). Elemental analysis for $C_{13}H_{16}N_4S$ (288.42); Calcd: C; 62.47, H; 6.99, N; 19.43, S; 11.12%. Found: C; 62.34, H; 7.55, N; 19.43, S; 10.71%.

Synthesis of 2,6-Dibenzyl-6,7-dihydro-5H-[1,2,4]triazolo[5,1-b]-[1,3,5]thiadiazines (6f)

R = $CH_2C_6H_5$. This was obtained as colorless crystals in 80% yield, m.p. 142°C, $R_f = 0.65$. IR (KBr) ν cm^{-1} = 3030 s (C—H aromatic), 2980 m, 2870 m (C—H aliphatic), 1600 m (C=N), 1580 s, 1520 s, 1460 s, (C=C aromatic skeleton), 1480 m, 1410 m,1220 m (C—H, C—N), 730 s, 700 s (aromatic). ^{1}H NMR ($CDCl_3$, 90 MHz) $\delta = 7.3$–7.1 (m, 10H, aromatic-H),

5.0 (s, 2H, NCH_2N), 4.5 (s, 2H, SCH_2N), 4.0 (s, 2H,$\mathbf{CH}_2$Ph), 3.8 (s, 2H, CH_2). The MS-EI m/z (%), $M^+ = 322$ (100%), 116 (34%), 91 (99%), 65 (18%), 203 (3%), 117 (10%), 145 (9%). Elemental analysis for $C_{18}H_{18}N_4S$ (322.43); Calcd: C; 67.05, H; 5.63, N; 17.38, S; 9.94%. Found: C; 67.06, H; 5.55, N; 17.29, S; 9.66%.

Synthesis of 2-Benzyl-6-(p-totyl)-6,7-dihydro-5H-[1,2,4]-triazolo[5,1-b][1,3,5]-thiadiazines (6g)

R = 4-$CH_3C_6H_4$. This was obtained as colorless crystals in 71% yield, m.p. 124°C, $R_f = 0.58$. IR (KBr) ν $cm^{-1} = 3030$ s (C—H aromatic), 2985 m, 2870 m (C—H aliphatic), 1610 m (C=N), 1580 s, 1510 s, 1460 s, (C=C aromatic skeleton), 1480 m, 1410 m,1220 m (C—H, C—N), 730 s, 700 s (aromatic). ^{1}H NMR ($CDCl_3$, 90 MHz) $\delta = 7.3$–6.7 (m, 9H, aromatic-H), 5.5 (s, 2H, NCH_2N), 4.9 (s, 2H, SCH_2N), 4.0 (s, 2H,$\mathbf{CH}_2$Ph), 2.2 (s, 3H, CH_3). The MS-EI m/z (%), $M^+ = 322$ (100%), 116 (34%), 91 (71%), 65 (25%), 203 (7%), 117 (22%), 145 (10%). Elemental analysis for $C_{18}H_{18}N_4S$ (322.43); Calcd: C; 67.05, H; 5.63, N; 17.38, S; 9.94%. Found: C; 67.32, H; 5.45, N; 17.27, S; 9.51%.

CONCLUSION

A new series of triazolothiadiazines **6a–g** were synthesized via double Mannich reaction in one pot synthesis in high yields. All synthesized compounds were screened for their antibacterial and antifungal activity in vitro. All tested compounds having an antifungal action at a dose of 1% concentration for fungi and bacteria.

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