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1,4-benzothiazepine Analogues: Synthesis, Characterisation and Antimicrobial Evaluation

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ABSTRACT

An efficient and accessible procedure for the synthesis of 1,4-benzothiazepines was developed. An acid catalyzed reaction of chalcones and 2-aminothiophenol produced 1,4-benzothiazepines in good yields. The synthesized new compounds were characterized by spectral studies and elemental analysis; and were screened *in vitro* for their antimicrobial potencies against different bacteria and fungi species.

Keywords: Antibacterial, Antifungal, Condensation, Inhibition, Spectral, Thiazepine

INTRODUCTION

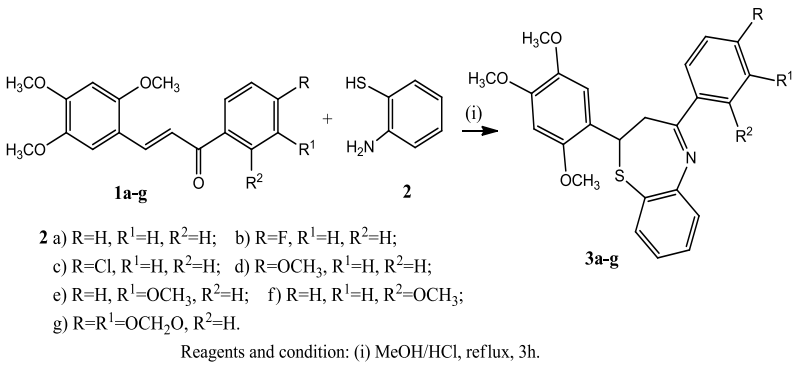
Chalcones forms the central core for the construction of variety of bioactive molecules such as benzothiazepine [1-4], pyrrolines [5], cyclopropyl esters [6], pyrazolines [7, 8], isoxazoles [9] etc. 1,4-Benzothiazepine skeleton is an important moiety that has been widely used as building block for pharmaceutical agents [10]. The broad spectrum of clinical importance and commercial success associated with benzothiazepines has led to their recognition in the medicinal chemistry [11]. The reaction of 2'-aminoethyl-3,4-dimethoxyphenyl sulfide hydrochloride with acid chlorides produced the acid amides, which on ring closure with phosphoryl chloride furnished the 2,3-dihydro-l,4-benzothiazepine [12,13]. (10-(4-Methylpiperazin-1-yl)pyrido[4,3-b][1,4]benzothiazepine) a structurally related to clozapine, was shown to be less sensitive to oxidation than clozapine. The effect of hypochlorous acid, appears that the oxidations of benzothiazepine derivative, unlike clozapine, are very slow and little secondary product does not cause the drug-induced agranulocytosis [14].

1, 4-Benzothiazepine derivatives are known to exhibit biological activities such as calcium channel blockers, antitumor agents, sedatives and hypnotics [15], antibiotics and anti histamine [16], antimicrobial [17] activity. Literature reveals that substituted benzothiazepines exhibited the strong antioxidant activity [18]. In view of broad spectrum of applications associated with 1,4-benzothiazepines, we herein report the synthesis and antimicrobial activity studies of series of new 1,4-benzothiazepine derivatives.

MATERIALS AND METHODS

Melting points were determined by an open capillary tube method and are uncorrected. Purity of the compounds was checked on Thin Layer Chromatography (TLC) plates pre-coated with silica gel using solvent system ethyl acetate: dichloromethane (1:4 v/v). The spots were visualized under UV light. Proton Nuclear Magnetic Resonance (^{1}H-NMR) and Carbon-13 Nuclear Magnetic Resonance (^{13}C-NMR) spectra were recorded on Agilent-NMR 400 MHz and 100 MHz spectrometer respectively. The solvent Deuterated Chloroform ($CDCl_3$) with Tetramethylsilane (TMS) as an internal standard was used to record the spectra. The chemical shifts are expressed in δ ppm. Mass spectra were obtained on Mass Lynx SCN781 spectrometer TOF mode. Elemental analysis was obtained on a Thermo Finnigan Flash EA 1112 CHN analyzer. Purification of compounds was done by column chromatography on silica gel (70-230 mesh Merck).

The required precursor chalcones, 1a-g was prepared according to our earlier reported procedure [19-22]. The cyclocondensation reaction of chalcones, 1a-g with 2-aminothiophenol, 2 in the presence of few drops of concentrated hydrochloric acid in methyl alcohol under reflux conditions produced derivatives, 3a-g in good yields. The schematic diagram for the synthesis of benzothiazepines is outlined in Figure 1.



2 a) R=H, R^1=H, R^2=H; b) R=F, R^1=H, R^2=H;
c) R=Cl, R^1=H, R^2=H; d) R=OCH_3, R^1=H, R^2=H;
e) R=H, R^1=OCH_3, R^2=H; f) R=H, R^1=H, R^2=OCH_3;
g) R=R^1=OCH_2O, R^2=H.

Reagents and condition: (i) MeOH/HCl, reflux, 3h.

Figure 1: Schematic diagram for the synthesis of benzothiazepines (3a-g)

General procedure for the synthesis of 1,4-benzothiazepines (3a-g)

To a stirred solution of chalcones, 1a-g (0.01 mol) and 2-aminothiophenol, 2 (0.01 mol) in methyl alcohol (15 ml), concentrated hydrochloric acid (7-8 drops) were added. The mixture was refluxed for 3-4 h and the progress of the reaction was monitored by TLC. After completion, the reaction mixture was poured in to ice cold water; solid separated was filtered, washed with ice cold water and dried. The products were purified column chromatography using silica gel (60-120 mesh) and ethyl acetate: n-hexane (1:3 v/v) as eluent.

4-Phenyl-2-(2,4,5-trimethoxyphenyl)-2,3-dihydrobenzo[b][1,4]thiazepine (3a)

Obtained from (E)-1-phenyl-3-(2,4,5-trimethoxyphenyl)prop-2-en-1-one, 1a (10 mmol) and 2-aminothiophenol, 2 (10 mmol) in 74% yield, m.p. 109-111°C; ^{1}H-NMR: δ=1.996-2.052 (dd, 1H, J=7.1Hz, 13.8Hz, C_3-H_a), 2.390-2.408 (dd, 1H, J=7.0Hz, 14.2Hz, C_3-H_b), 3.853 (s, 9H, OCH_3), 4.196-4.224 (dd, 1H, J=8.6Hz, 16.3Hz, C_2-H), 7.210–7.948 (m, 11H, Ar–H); ^{13}C-NMR: δ=41.23 (1C, C-3), 43.88 (1C, C-2), 55.55 (3C), 100.10 (1C), 108.12 (1C), 114.26 (1C), 117.66 (1C), 125.46 (1C), 127.18 (1C), 128.40 (2C), 128.94 (2C), 131.02 (1C), 133.50 (1C), 137.24 (1C), 137.97 (1C), 142.25 (1C), 148.78 (1C), 151.14 (1C), 155.44 (1C), 161.89 (1C, C-4); MS *m/z*: 405 (M^+, 100); Anal. Calcd. For $C_{24}H_{23}NO_3S$ (%): C, 71.09; H, 5.72; N, 3.45; Found: C, 71.00; H, 5.56; N, 3.33.

4-(4-Fluorophenyl)-2-(2,4,5-trimethoxyphenyl)-2,3-dihydrobenzo[b][1,4]thiazepine (3b)

Obtained from (E)-1-(4-fluorophenyl)-3-(2,4,5-trimethoxyphenyl)prop-2-en-1-one, 1b (10 mmol) and 2-aminothiophenol, 2 (10 mmol) in 78% yield; ^{1}H-NMR: δ=1.982-2.102 (dd, 1H, J=7.8Hz, 13.5Hz, C_3-H_a), 2.3860-2.410 (dd, 1H, J=7.3Hz, 14.0Hz, C_3-H_b), 3.855 (s, 9H, OCH_3), 4.1886-4.202 (dd, 1H, J=8.0Hz, 15.1Hz, C_2-H), 7.256–7.882 (m, 10H, Ar–H); ^{13}C-NMR: δ=41.26 (1C, C-3), 43.97 (1C, C-2), 55.58 (3C), 100.42 (1C), 107.48 (1C), 114.02 (1C), 115.56 (2C), 117.65 (1C), 125.61 (1C), 127.50 (1C), 129.10 (2C), 133.48 (1C), 133.62 (1C), 137.45 (1C), 142.40 (1C), 148.92 (1C), 150.77 (1C), 155.88 (1C), 164.40 (1C); MS *m/z*: 423 (M^+, 100); Anal. Calcd. For $C_{24}H_{22}FNO_3S$ (%): C, 68.07; H, 5.24; N, 3.31; Found: C, 68.01; H, 5.12; N, 3.24.

4-(4-Chlorophenyl)-2-(2,4,5-trimethoxyphenyl)-2,3-dihydrobenzo[b][1,4]thiazepine (3c)

Obtained from (E)-1-(4-chlorophenyl)-3-(2,4,5-trimethoxyphenyl)prop-2-en-1-one, 1c (10 mmol) and 2-aminothiophenol, 2 (10 mmol) in 81% yield. ^{1}H-NMR: δ=1.976-2.120 (dd, 1H, J=6.8Hz, 13.0Hz, C_3-H_a), 2.360-2.402 (dd, 1H, J=7.3Hz, 14.0Hz, C_3-H_b), 3.850 (s, 9H, OCH_3), 4.177-4.213 (dd, 1H, J=8.3Hz, 15.1Hz, C_2-H), 7.233–7.850 (m, 10H, Ar–H); ^{13}C-NMR: δ=40.54 (1C, C-3), 43.44 (1C, C-2), 55.60 (3C), 100.55 (1C), 107.88 (1C), 113.98 (1C), 117.20 (1C), 125.20 (1C), 127.25 (1C), 128.52 (2C), 128.96 (2C), 133.52 (1C), 135.10 (1C), 136.50 (1C), 138.10 (1C), 143.07 (1C), 148.20 (1C), 150.41 (1C), 155.33 (1C), 161.81 (1C, C-4); MS *m/z*: 441 (M^+, ^{37}Cl, 34), 439 (M^+, ^{35}Cl, 100); Anal. Calcd. For $C_{24}H_{22}ClNO_3S$ (%): C, 65.52; H, 5.04; N, 3.18; Found: C, 65.40; H, 5.00; N, 3.12.

4-(4-Methoxyphenyl)-2-(2,4,5-trimethoxyphenyl)-2,3-dihydrobenzo[b][1,4]thiazepine (3d)

Obtained from (E)-1-(4-methoxyphenyl)-3-(2,4,5-trimethoxyphenyl)prop-2-en-1-one, 1d (10 mmol) and 2-aminothiophenol, 2 (10 mmol) in 84% yield; ^{1}H-NMR: δ=1.980-2.041 (dd, 1H, J=7.3, 13.6Hz, C_3-H_a), 2.386-2.414 (dd, 1H, J=7.9, 14.5Hz, C_3-H_b), 3.856 (s, 12H, OCH_3), 4.206-4.220 (dd, 1H, J=8.7, 16.0Hz, C_2-H), 7.245–7.966 (m, 10H, Ar–H); ^{13}C-NMR: δ=40.98 (1C, C-3), 43.92 (1C, C-2), 55.66 (4C), 100.10 (1C), 107.25 (1C), 113.20 (1C), 114.42 (2C), 117.22 (1C), 125.40 (1C), 127.20 (1C), 128.90 (2C), 129.67 (1C), 133.56 (1C), 137.14 (1C), 143.74 (1C), 148.30 (1C), 151.44 (1C), 155.50 (1C), 161.98 (1C, C-4), 163.50 (1C); MS *m/z*: 435 (M^+, 100); Anal. Calcd. For $C_{25}H_{25}NO_4S$ (%): C, 68.94; H, 5.79; N, 3.22; Found: C, 68.82; H, 5.68; N, 3.11.

4-(3-Methoxyphenyl)-2-(2,4,5-trimethoxyphenyl)-2,3-dihydrobenzo[b][1,4]thiazepine (3e)

Obtained from (E)-1-(3-methoxyphenyl)-3-(2,4,5-trimethoxyphenyl)prop-2-en-1-one, 1e (10 mmol) and 2-aminothiophenol, 2 (10 mmol) in 80% yield; ^{1}H-NMR: δ=1.978-2.102 (dd, 1H, J=7.6, 13.5Hz, C_3-H_a), 2.390-2.422 (dd, 1H, J=7.0, 13.2Hz, C_3-H_b), 3.851 (s, 12H, OCH_3), 4.212-4.236 (dd, 1H, J=8.8, 15.3Hz, C_2-H), 7.233–7.945 (m, 10H, Ar–H); ^{13}C-NMR: δ=40.22 (1C, C-3), 43.76 (1C, C-2), 55.48 (4C), 100.34 (1C), 107.44 (1C), 113.56 (1C), 114.55 (2C), 117.40 (1C), 125.53 (1C), 127.34 (1C), 128.66 (2C), 129.60 (1C), 133.52 (1C), 137.10 (1C), 143.85 (1C), 148.90 (1C), 151.33 (1C), 155.67 (1C), 161.95 (1C, C-4), 163.46 (1C); MS *m/z*: 435 (M^+, 100); Anal. Calcd. For $C_{25}H_{25}NO_4S$ (%): C, 68.94; H, 5.79; N, 3.22; Found: C, 68.80; H, 5.65; N, 3.14.

4-(2-Methoxyphenyl)-2-(2,4,5-trimethoxyphenyl)-2,3-dihydrobenzo[b][1,4]thiazepine (3f)

Obtained from (E)-1-(2-methoxyphenyl)-3-(2,4,5-trimethoxyphenyl)prop-2-en-1-one, 1f (10 mmol) and 2-aminothiophenol, 2 (10 mmol) in 75% yield; ^{1}H-NMR: δ=1.987-2.124 (dd, 1H, J=7.0, 13.3Hz, C_3-H_a), 2.390-2.423 (dd, 1H, J=7.4, 14.9Hz, C_3-H_b), 3.854 (s, 12H, OCH_3), 4.212-4.234 (dd, 1H, J=8.1, 16.7Hz, C_2-H), 7.240–7.972 (m, 10H, Ar–H); ^{13}C-NMR: δ=41.12 (1C, C-3), 43.34 (1C, C-2), 55.45 (4C), 100.78 (1C), 107.30 (1C), 113.46 (1C), 114.34 (2C), 117.55 (1C), 125.67 (1C), 127.10 (1C), 128.80 (2C), 129.62 (1C), 133.50 (1C), 137.25 (1C), 143.64 (1C), 148.60 (1C), 151.77 (1C), 155.80 (1C), 161.90 (1C, C-4), 163.57 (1C); MS *m/z*: 435 (M^+, 100); Anal. Calcd. For $C_{25}H_{25}NO_4S$ (%): C, 68.94; H, 5.79; N, 3.22; Found: C, 68.79; H, 5.63; N, 3.09.

4-(Benzo[d][1,3]dioxol-5-yl)-2-(2,4,5-trimethoxyphenyl)-2,3-dihydrobenzo[b][1,4]thiazepine (3g)

Obtained from (E)-1-(benzo[d][1,3]dioxol-5-yl)-3-(2,4,5-trimethoxyphenyl)prop-2-en-1-one, 1 g (10 mmol) and 2-aminothiophenol, 2 (10 mmol) in 88% yield; ^{1}H-NMR: δ=1.990-2.088 (dd, 1H, J=7.0Hz, 14.5Hz, C_3-H_a), 2.387-2.412 (dd, 1H, J=6.7Hz, 14.0Hz, C_3-H_b), 3.855 (s, 9H, OCH_3), 4.198-4.218 (dd, 1H, J=8.2Hz, 16.1Hz, C_2-H), 6.051 (s, 2H, OCH_2O), 7.156–7.966 (m, 9H, Ar–H); ^{13}C-NMR: δ=41.35 (1C, C-3), 44.30 (1C, C-2), 55.50 (3C), 100.87 (1C), 102.86 (1C), 109.26 (1C), 111.16 (1C), 113.87 (1C), 114.40 (1C), 117.10 (1C), 121.10 (1C), 125.45 (1C), 127.33 (1C), 127.76 (1C), 133.54 (1C), 137.10 (1C), 142.50 (1C), 148.36 (1C), 149.30 (1C), 150.64 (1C), 151.40 (1C), 155.46 (1C), 161.42 (1C, C-4), 163.52 (1C); MS *m/z*: 449 (M^+, 100); Anal. Calcd. For $C_{25}H_{23}NO_5S$ (%): C, 66.80; H, 5.16; N, 3.12; Found: C, 66.71; H, 5.10; N, 3.02.

RESULTS AND DISCUSSION

Structure proof of synthesized compounds, 3a-g were provided by ^{1}H-NMR, ^{13}C-NMR, mass spectral studies and elemental analysis. The structural assignments were made by spectral analysis by considering 4-(4-chlorophenyl)-2-(2,4,5-trimethoxyphenyl)-2,3-dihydrobenzo[b][1,4]thiazepine, 3c as the representative compound among the series. In ^{1}H-NMR spectra, 2 methylene protons designated as C_3-H_a and C_3-H_b of the newly formed pyrazoline ring is diastereotopic. The C_3-H_a, C_3-H_b and C_2-H protons appeared as a doublet of doublets. The doublet of doublet for C_3-H_a appeared in the region δ=1.976-2.120 (J=6.8, 13.0Hz) ppm; doublet of doublet for C_3-H_b appeared in the region δ=2.360-2.402 (J=7.3, 14.0 Hz) ppm; and that of C_2-H in the region δ=4.177-4.213 (J=8.3, 15.1Hz) ppm. Among C_3-H_a, C_3-H_b and C_2-H protons, C_2-H is the most deshielded due to its close proximity to aromatic ring and electronegative nitrogen. C_2-H couples not only with C_3-H_a but also with C_3-H_b and appears as doublet of doublet instead of a triplet. A collection of signal observed singlet for eleven protons at δ=3.850 ppm and as multiplet for twelve protons in the region δ=7.233-7.850 ppm were assigned to OCH_3 and aromatic protons respectively.

In ^{13}C-NMR spectrum, compound 3c showed a signal at δ=40.54, 43.44 and 161.81 ppm due to C-3, C-2 and C-4 carbons of the benzothiazepine ring. A signal appeared for three carbons at δ=55.60 ppm was assigned to 3-OCH_3 carbons. An array of signals appeared at δ=100.55, 107.88, 113.98, 117.20, 125.20, 127.25, 128.52, 128.96, 133.52, 135.10, 136.50, 138.10, 143.07, 148.20, 150.41 and 155.33 ppm were assigned to aromatic carbons. Compound 3c showed molecular ion peaks at m/z 441 (M^+, ^{37}Cl, 34) and m/z 439 (M^+, ^{35}Cl, 100) corresponding to its molecular mass. Satisfactory analytical data obtained for the compound further supports the structure of the compound. The synthesized series of compounds showed similar and consistent pattern signals in their respective spectra, which strongly favors the structure proof for the synthesized compounds.

Antimicrobial activity

Antimicrobial studies of synthesized compounds 3a-g were assessed by Minimum Inhibitory Concentration (MIC) by serial dilution method [23]. The compounds were screened for their antimicrobial activities against Gram-negative bacteria *Escherichia coli*, Gram-positive bacteria *Staphylococcus aureus*, fungi species *Aspergillus nigar* and *Aspergillus flavus.* The experiments were carried out in triplicate; the results were taken as a mean of three determinations. The antibiotics ciprofloxacin and nystatin were used as standard drugs for antibacterial and antifungal studies respectively. The results of MIC's were tabulated in Table 1.

Table 1: Antimicrobial activities of the compounds 3a-g against bacterial and fungal stains

Compound	Minimum Inhibitory Concentration (MIC's) in µg/ml*			
	Staphylococcus aureus	***Escherichia coli***	***Aspergillus niger***	***Aspergillus flavus***
3a	25	12.5	12.5	25
3b	25	12.5	12.5	25
3c	12.5	12.5	12.5	12.5
3d	125	150	125	150
3e	150	150	125	150
3f	150	150	125	150
3g	50	25	25	50
Ciprofloxacin	25	12.5	-	-
Nystatin	-	-	12.5	25

*The results are expressed as mean of three determinations (n=3)

Preliminary investigation results of antimicrobial evaluation of the synthesized benzothiazipine derivatives 3a-g reveals that these compounds exert varied antimicrobial susceptibilities against all the tested organisms. Among the synthesized series, in comparison with the standards, compounds 3a, 3b and 3c having unsubstitution, fluoro and chloro substitution on the C-4 substituted aromatic ring exhibited an excellent activities against all the tested species. Compound 3g with methylenedioxy substitution found good active; while the compounds 3d, 3e and 3f with methoxy substitutions in the aromatic ring lesser inhibition potential against the testes organisms.

CONCLUSION

The reaction described represents a simple accessible route for the synthesis of 1,4-benzothiazepine derivatives. Preliminary studies on the antimicrobial activities of the synthesized 1,4-benzothiazepines reveals that some of the synthesized series of compounds acts as a potential antimicrobial agents.

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