Synthesis and Antimicrobial Studies of Novel Bis(diamino)thiazoles

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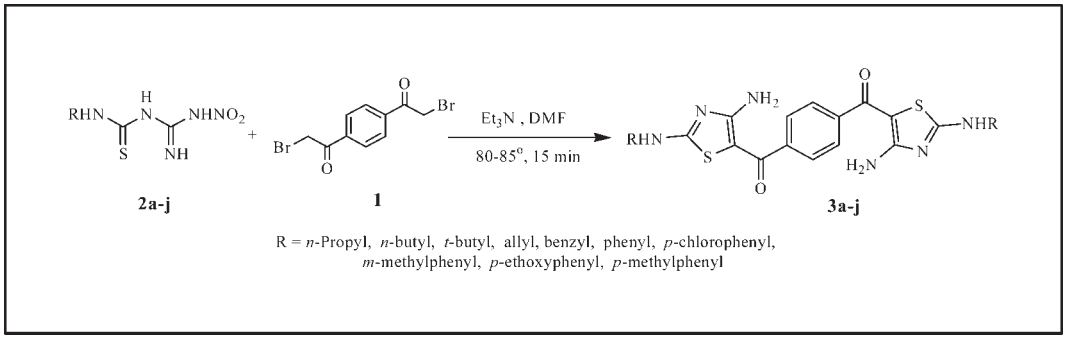

The synthesis of novel bis(diamino)thiazoles has been achieved by reacting bis(bromoacetyl)benzene and 1-alkyl(or aryl)-3-(*N*-nitroamidino)thioureas in presence of triethylamine. These new compounds were characterized by spectral analysis and screened for antimicrobial activities.

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INTRODUCTION

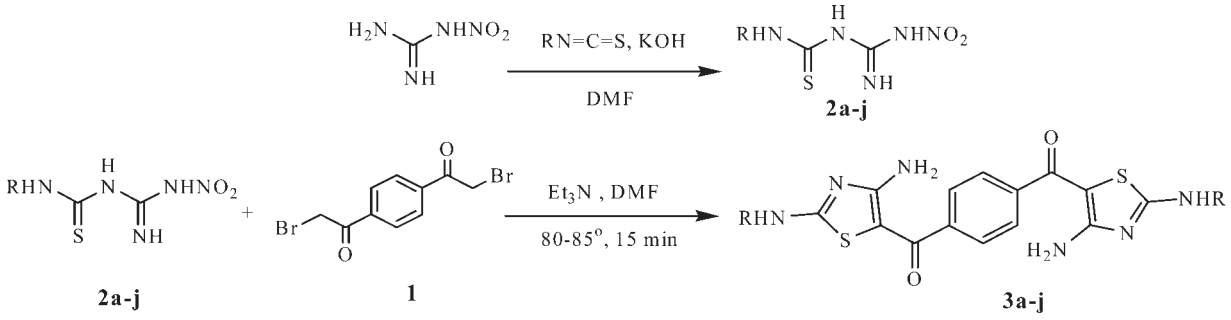
Compounds that incorporate two thiazole rings either directly connected as in bithiazoles or through a linker unit as in bisthiazolyl compounds, show significant biological activity. Bleomycin, isolated from *Streptomyces verticilus* containing a bithiazole moiety, which has been shown to bind to DNA thereby inhibiting DNA synthesis of the tumor cells, is the prime example for such compounds [1]. A number of other bi- or bisthiazoles [2–4] exhibit antibacterial, antiviral and cytotoxic activities. In addition to these compounds, quite a few of the highly active secondary metabolites from marine tunicates also contain bisthiazole units [5]. It also appears that the thiazole units need not always be directly linked for the manifestation of bioactivity; for example, ritonavir, a bisthiazole in which the two thiazole units are separated by a small peptidomimetic chain, is a potent bisthiazole HIV-protease inhibitor [6].

As far back as in 1997, we have observed that 2,4-diamino-5-ketothiazoles show excellent cytotoxic activity [7] based on screening programs at National Cancer Institute, an observation later substantiated further by us [8]. As part of our work in the synthesis of 2,4-diamino-5-ketothiazoles, we have developed a variety of thiourea based synthons, such as amidinothioureas, nitroamidinothioureas, and thiocarbamoylamidinopyrazoles that [9] serve as precursors for 2,4-diaminothiazoles by providing the [C^4—N^3—C^2—S^1] ring atoms for the thiazole ring construction, the remaining C^5 carbon of the thiazole being sourced from a reactive halomethyl compound of the type X—CH_2—EWG where the electron withdrawing group EWG can be O_2N—C_6H_4—, NC—, R—CO—, RO—CO—, Ar—CH=CH—CO— or similar ones, including α-haloketones. This [4 + 1] ring assembly, in a tandem bromination-cyclization strategy has been now applied for the synthesis of novel [4-(4-amino-2-(*N*-substituted amino)thiazole-5-carbonyl)-phenyl]-(4-amino-2-(*N*-substituted amino)thiazol-5-yl)-methanones. The above [(C^4—N^3—C^2—S^1) + C^5] ring assembly of 2,4-diaminothiazole using functionalized thiourea derivatives is distinctly different from the classic Hantzsch synthesis, which is the synthesis of 2-aminothiazoles from halo carbonyl compounds and thioureas is well reviewed [10,11] in literature. The latter is a [3 + 2] ring construction strategy starting from simple thioureas, that give the [N^3—C^2—S^1] ring atoms, and α-haloketones, which provide the remaining [C^4—C^5] atoms, giving 2-aminothiazoles as products.

RESULTS AND DISCUSSION

In a typical case, the reaction of 1-(*N*-nitroamidino)-3-*n*-propylthiourea with 1,4-bis(bromoacetyl)benzene

Scheme 1



R = *n*-Propyl, *n*-butyl, *t*-butyl, allyl, benzyl, phenyl, *p*-chlorophenyl, *m*-methylphenyl, *p*-ethoxyphenyl, *p*-methylphenyl

was done in DMF in the presence of triethylamine at 80–85°C for 15 min to obtain a compound which had a molecular composition $C_{20}H_{24}N_6O_2S_2$ gave a product the FAB mass spectrum of the which showed a MH^+ peak at *m*/*z* 445 indicating that the molecular mass of the compound is 444. It's ^1H NMR spectrum showed the presence of an *n*-propylamino group, a phenylene group and NH hydrogen. The ^{13}C NMR spectrum showed three peaks in the alkyl region at 13.51, 20.12, and 31.50 ppm indicating the presence of the *n*-propyl group. The signals at 143.8, 156.7, and 167 ppm were assignable to thiazole ring carbons, that at 181 ppm to carbonyl carbon and those at 126.4, 127.6, 128, and 129 ppm, to a *p*-phenylene group carbons. Accordingly, the compound was formulated as [4-(4-amino-2-*n*-propylaminothiazole-5-carbonyl)phenyl]-(4-amino-2-*n*-propylaminothiazol-5-yl)methanone **3a**. The reaction was next extended to 1-aryl-3-(*N*-nitroamidino)thioureas. As a typical example, 1-(4-methylphenyl-3-(*N*-nitroamidino)thiourea reacted with 1,4-bis(bromoacetyl)benzene to give a compound with molecular composition $C_{28}H_{24}N_6O_2S_2$. The ^1H NMR spectrum of the compound showed a doublet due to four hydrogens at δ 7.02–7.25, assignable to aryl hydrogen *ortho*- to a NH group. Another doublet of four hydrogens at δ 7.35–7.55 was ascribed to the aryl hydrogen *meta*- to a NH group. A singlet of four hydrogens at δ 7.74 was attributable to the aryl hydrogen of a *p*-phenylene group. A broad peak at δ 8.0–8.52 was due to NH hydrogen. A singlet due to two hydrogens at δ 10.72 was also assignable to two NH groups. The ^{13}C NMR spectrum showed fourteen peaks among which the peak at δ 26.98 showed the presence of a methyl group. Based on these data and the fragmentations observed in its EI mass spectrum, the compound was assigned the structure [4-(4-amino-4-phenylaminothiazole-5-carbonyl)phenyl]-(4-amino-4-phenylaminothiazol-5-yl)methanone **3j** (Scheme 1).

The general reaction scheme is depicted above. These new [4-(4-amino-2-(*N*-substituted amino)thiazole-5-carbonyl)phenyl]-(4-amino-2-(*N*-substituted amino)thiazol-5-yl)methanones (**3a–j**) were screened for their antibacterial and antifungal activities. The data from these studies are compiled in Table 1.

In conclusion, we have described an efficient synthetic route to obtain bis(diamino)thiazoles (**3a–j**), several of which show good antibacterial and antifungal activities.

EXPERIMENTAL

Melting points were determined by open capillary method and were uncorrected. The IR spectra were recorded in potassium bromide pellets with an AVATAR 330 FTIR spectrometer and the ^1H, ^{13}C NMR experiments were done on AMX-400/DRX-500 NMR spectrometers. The molecular masses of compounds were confirmed by taking EI and the fast atom bombardment mass spectra (FAB-MS). The compounds were purified by column chromatography using silica gel (60–120 mesh, E. Merck). The required alkyl and aryl isothiocyanates and nitroamidinothioureas were synthesized by reported methods [9(c),12] The bromination of 2,4-diacetylbenzene was done in DMF at 80–85°C and the 1,4-bis(bromoacetyl)benzene **1** was isolated and characterized. The antibacterial and antifungal activities were studied by the disc diffusion method against *Staphylococcus aureus*, *Klebsiella pneumonia*, *Candida albicans*, and *Aspergillus niger*. In the case of *C. albicans* and *A. niger* the standard used is ketoconazole and in the case of *S. aureus* and *K. pneumonia* the data has been compared with ciplofloxacin.

Bis(bromoacetyl)benzene (1). To a solution of 1,4-diacetylbenzene (0.21 g, 1.3 mmol) in glacial acetic acid, bromine (2.6 mmol, 90 mL) in glacial acetic acid (2 mL) was added slowly and warmed. The reaction mixture was then heated at 95°C for 1 h, and then poured into ice water and the product was filtered, dried and recrystallized from glacial acetic, to obtain colorless shinning crystals of 1,4-bis(bromoacetyl)benzene (**1**) in 65% yield, mp 165–167°C; IR (KBr): 2998, 2945, 1699, 1402, 1263, 1202, 1121, 987, 811, 683, 565, 499 cm^{-1}; ^1H NMR (400 MHz, $CDCl_3$): δ: 4.45 (s, 4H, CH_2), 8.04–8.10 (m, 4H, Ar—H); FAB ms: *m*/*z* (MH^+) 320.

1-*t*-Butyl-3-(*N*-nitroamidino)thiourea (2c). Finely powdered potassium hydroxide (1.08 g, 1.92 mmol) was added to

Table 1

Antimicrobial activity studies of compounds (**3a-j**) by disc diffusion method.

	Zone of inhibition (mm)							
	Bacterial strain				Fungal strain			
	S. aureus		*K. pneumonia*		*C. albicans*		*A. niger*	
Compound	Std	Sample	Std	Sample	Std	Sample	Std	Sample
3a	29	17	28	19	29	19	26	13
3b	27	22	33	19	33	22	24	19
3c	28	18	31	22	32	18	23	19
3d	22	21	30	20	31	19	19	16
3e	26	18	32	20	33	21	23	19
3f	28	23	33	17	31	20	21	13
3g	27	20	32	23	32	19	20	17
3h	29	20	32	27	33	22	24	19
3i	27	24	32	22	32	19	22	17
3j	29	19	32	26	33	21	23	18

Concentration: l00 μg/mL.

a solution of nitroguanidine (1 g, 9.6 mmol) in *N*,*N*-dimethylformamide (10 mL) and this mixture was stirred for 15 min. To this, *t*-butyl isothiocyanate (9.6 mmol) in *N*,*N*-dimethylformamide (5 mL) was added in 15 min and the stirring was continued for another 60 min. The reaction mixture was then poured onto crushed ice and the mixture was acidified with dilute hydrochloric acid (0.1*N*). The precipitated compound was filtered, dried and crystallized from ethanol to obtain 1-*t*-butyl-3-(*N*-nitroamidino)thiourea (**2c**) in 71% yield, mp 146–148°C; IR (KBr) 3353, 3284, 2983, 2934, 2545, 1715, 1647, 1608, 1545, 1365, 1259,1191,1076, 957, 786, 705, 632 cm^{-1},^{1}H NMR (400 MHz, DMSO-d_6): δ 1.4 (s, 9H, CH_3), 9.10–9.45 (br s, 1H, NH), 9.66 (s, 1H, NH), 9.96 (s, 1H, NH); FAB ms: *m*/*z* (MH^+) 220. *Anal*. Calcd. for $C_6H_{13}N_5O_2S$: C, 32.86; H, 5.98; N, 31.94%. Found: C, 33.00; H, 6.09; N, 31.81%.

General procedure for the synthesis of bis(diamino)thiazoles (3a–j). To a solution of 1,4-bis(bromoacetyl)benzene in DMF (2 mL) kept at 50–60°C containing triethylamine (2 mmol), 1-alkyl/aryl-3-(*N*-nitroamidino)thioureas (1 mmol), in *N*,*N*-dimethylformamide (2 mL) was added dropwise. After warming for 15 min, the reaction mixture was poured into water. The product obtained, after adjusting the pH to 7, was collected and purified using column chromatography using silica gel by elution using chloroform-ethyl acetate (1:5) to obtain the bisthiazoles in 65–80% yield.

[4-(4-Amino-2-n-propylaminothiazole-5-carbonyl)phenyl]-(4-amino-2-n-propylaminothiazol-5-yl)-methanone (3a). This compound was obtained as yellow-colored microcrystals (65%), mp > 250°C; IR (KBr): 3416, 2963, 2928, 1603, 1567, 1444, 1384, 1279, 1225, 1091, 747 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ 0.85 (t, 6H, CH_3, J = 7 Hz), 1.53 (sextet, 4H, CH_2, J = 7 Hz), 3.68 (t, 4H, CH_2, J = 5.1 Hz), 7.60–8.10 (m, 4H, Ar—H), 8.69 (s, NH); ^{13}C NMR (100 MHz, DMSO-d_6)δ: 13.51, 20.12, 31.50, 126.4, 127.6, 128, 129, 143.8, 156.7, 167, 181 ppm; FAB ms: *m*/*z* (MH^+) 445. *Anal*. Calcd. for $C_{20}H_{24}N_6O_2S_2$: C, 54.03; H, 5.44; N, 18.91%. Found: C, 54.00; H, 5.40; N, 18.81%.

[4-(4-Amino-2-n-butylaminothiazole-5-carbonyl)phenyl]-(4-amino-2-n-butylaminothiazol-5-yl)methanone (3b). This compound was obtained as yellow-colored microcrystals (60%), mp > 250°C; IR (KBr): 3347, 2958, 2926, 2857, 1703, 1592, 1518, 1463, 1407, 1261, 1229, 1097, 801, 732, 567 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6): δ 0.72 (t, 6H, CH_3),1.32 (m, 4H, CH_2, J = 7 Hz), 1.40–1.60 (q, 4H, CH_2), 3.72 (t, 4H, CH_2), 7.40–8.30 (m, 4H, Ar—H), 8.6 (br s, 1H, NH); ^{13}C NMR (100 MHz, DMSO-d_6): δ 12.11, 13.53, 19.46, 30.52, 126.64, 126.92, 128.55, 129.30, 143.8, 156.66, 168, 179, 180ppm; FAB ms: *m*/*z* (MH^+) 473. *Anal*. Calcd. for $C_{22}H_{28}N_6O_2S_2$: C, 55.91; H, 5.97; N, 17.78%. Found: C, 55.85; H, 5.92; N, 17.69%.

[4-(4-Amino-2-t-butylaminothiazole-5-carbonyl)phenyl]-(4-amino-2-t-butylaminothiazol-5-yl)methanone (3c). This compound was obtained as yellow-colored microcrystals (55%), mp > 250°C; IR (KBr): 3281, 2970, 2929, 1685, 1603, 1557, 1437, 1367, 1275, 1200, 1096, 905, 851, 749 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6): δ 1.35 (s, 18H, CH_3), 7.6–8.5 (m, 4H, Ar—H); ^{13}C NMR (100 MHz, DMSO-d_6): δ 27.34, 28.17, 29.05, 52.39, 126.40, 126.60, 128.04, 129.31, 143.28, 156.31, 166.32, 169.27, 179.74, 180.41 ppm; FAB ms: *m*/*z* (MH^+) 473. *Anal*. Calcd. for: Found: C, 55.89; H, 5.91; N, 17.82%; Calculated for $C_{22}H_{28}N_6O_2S_2$: C, 55.91; H, 5.97; N, 17.78%.

[4-(2-Allylamino-4-aminothiazole-5-carbonyl)phenyl]-(2-allylamino-4-aminothiazol-5-yl)methanone (3d). This compound was obtained as yellow-colored microcrystals (48%), mp > 250°C; IR (KBr): 3440, 2925, 1608, 1566, 1446, 1384, 1309, 1226, 1091, 929, 581 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6): δ 3.70–4.10 (m, 4H, CH_2), 4.9–5.4 (m, 4H, CH_2), 5.70–6.00 (m, 2H, CH), 7.6–8.2 (m, 4H, ArH); ^{13}C NMR (100 MHz, DMSO-d_6): δ 28.81, 46.8, 54.8, 72.15, 92.6, 116.2, 118.37, 166.6, 180.1, 193.6 ppm; FAB ms: *m*/*z* (MH^+) 441. *Anal*. Calcd. for $C_{20}H_{20}N_6O_2S_2$: C, 54.52; H, 4.58; N, 19.08%. Found: C, 54.43; H, 4.44; N, 19.18%

[4-(4-amino-2-n-benzylaminothiazole-5-carbonyl)phenyl]-(4-amino-2-n-benzylaminothiazol-5-yl)methanone (3e). This compound was obtained as yellow-colored microcrystals (51%), mp > 250°C; IR (KBr): 3409, 2958, 2927, 1703, 1581, 1518, 1464, 1407, 1230, 1103 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6): δ 4.5 (s, 2H, CH_2), 7.1–8.1 (m, Ar—H); ^{13}C NMR (100 MHz, DMSO-d_6): δ 47, 48, 126, 127, 128, 129, 137.7; FAB ms: *m*/*z* (MH^+) 541. *Anal*. Calcd. for $C_{28}H_{24}N_6O_2S_2$: C,

62.20; H, 4.47; N, 15.55%. Found: C, 62.28; H, 4.54; N, 15.32%.

[4-(4-Amino-2-phenylaminothiazole-5-carbonyl)phenyl]-(4-amino-2-phenylaminothiazol-5-yl)methanone (3f). This compound was obtained as yellow-colored microcrystals (42%), mp > 250°C; IR (KBr): 3435, 2360, 1659, 1598, 1549, 1440, 890, 853, 780, 743, 698 cm^{-1};^{1}H NMR (400 MHz, DMSO-d_6): δ 7.05 (t, 2H, 2Ar—H), 7.31(t, 4H, 4Ar—H), 7.58 (d, 4H, 4Ar—H), 7.67–7.81(m, 8H, 4Ar—H, $2NH_2$); ^{13}C NMR (100 MHz, DMSO-d_6): δ 93.14, 119.51, 123.66, 127.18, 129.27, 139.93, 143.74, 166.38, 167.92, 182.16 ppm; FAB ms: *m/z* (MH^+) 513; *Anal*. Calcd. for $C_{26}H_{20}N_6O_2S_2$: C, 60.92; H, 3.93; N, 16.40%. Found: C, 60.75; H, 3.81; N, 16.30%.

{4-[4-Amino-2-(4-chlorophenylamino)thiazole-5-carbonyl]-phenyl}-[4-amino-2-(4-chlorophenylamino)thiazol-5-yl]methanone (3g). This compound was obtained as yellow-colored microcrystals (42%), mp > 250°C; IR (KBr): 3430, 2360, 1598, 1530, 1492, 1426, 1091, 825, 668 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6):δ 6.75–8.1 (m, 16H), 10.15 (s, 2H, NH); EI ms: *m/z* (%) 453 (2), 411 (2), 224 (11),184 (4), 169 (26), 153 (11), 152 (20), 127 (100), 111 (48). *Anal*. Calcd. for $C_{26}H_{18}Cl_2N_6O_2S_2$: C, 53.70; H, 3.12; N, 14.45%. Found: C, 53.55; H, 3.09; N, 14.32%.

{4-[4-Amino-2-(3-methylphenylamino)thiazole-5-carbonyl]-phenyl}-[4-amino-2-(3-methylphenylamino)thiazol-5-yl]methanone (3h). This compound was obtained as yellow-colored microcrystals (58%), mp > 250°C; IR (KBr): 3440, 3220, 2361, 1552, 1422, 1167, 1088, 898, 850, 781, 745, 693 cm^{-1}; ^{1}H NMR (250 MHz, DMSO-d_6): δ 2.65 (s, 6H, CH_3), 6.98–7.2 (d, 4H, J = 6.38 Hz), 7.29–7.45 (d, 4H, J = 6.43 Hz), 7.75 (s, 4H); ^{13}C NMR (100 MHz, DMSO-d_6): δ 26.82, 94.47, 117.34, 120.97, 125.49, 127.44, 128.34, 129.16, 138.07, 138.90, 139.34, 145.83, 165.87, 169.64, 182.80 ppm; FAB ms: *m/z* (MH^+) 541. *Anal*. Calcd. for $C_{28}H_{24}N_6O_2S_2$: C, 62.22; H, 4.47; N, 15.55%. Found: C, 62.17; H, 4.52; N, 15.29%.

{4-[4-Amino-2-(4-ethoxyphenylamino)thiazole-5-carbonyl]-phenyl}-[4-amino-2-(4-ethoxyphenylamino)thiazol-5-yl]methanone (3i). This compound was obtained as yellow-colored microcrystals (35%), mp > 250°C; IR (KBr): 3483, 3311, 3306, 3275, 2996, 1591, 1510, 1428, 1235, 1180, 1092, 1035, 822, 746 cm^{-1}; ^{1}H NMR (400 MHz, DMSO-d_6): δ 1.39 (t, 6H, CH_3), 4.02 (q, 4H, CH_2), 6.86 (d, Ar—H), 7.30–7.50 (m, Ar—H), 7.73 (s, Ar—H), 7.94 (s, Ar—H), 10.40 (s, NH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ 14.30, 62.83, 114.24, 121.16, 126.34 ppm; FAB ms: *m/z* (MH^+) 601. *Anal*. Calcd. for $C_{30}H_{28}N_6O_4S_2$: C, 59.98; H, 4.70; N, 13.99%. Found: C, 59.78; H, 4.61; N, 13.73%.

{4-[4-Amino-2-(4-methylphenylamino)thiazole-5-carbonyl]-phenyl}-[4-amino-2-(4-methylphenylamino)thiazol-5-yl]methanone (3j). This compound was obtained as yellow-colored microcrystals (52%), mp > 250°C; IR (KBr): 3445, 3221, 2358, 1551, 1423, 1167, 1076, 892, 850, 770, 741, 690 cm^{-1}; ^{1}H NMR (300 MHz, DMSO-d_6): δ 2.60 (s, 6H, CH_3), 7.02–7.25 (d, 4H, J = 6.49 Hz), 7.35–7.55 (d, 4H, J = 6.52 Hz), 7.74 (s, 4H), 8.0–8.52 (br, 4H, NH_2), 10.72 (s, 2H), ^{13}C NMR (100 MHz, DMSO-d_6): δ 26.98, 93.76, 117.98, 119.76, 123.91, 127.35, 128.41, 129.21, 138.01, 139.59, 146.05, 166.50, 168.65, 182.03 ppm; El ms: *m/z* (%) 149 (34), 147 (8), 133 (10), 107 (64), 106 (38), 91 (100). *Anal*. Calcd. for $C_{28}H_{24}N_6O_2S_2$: C, 62.22; H, 4.47; N, 15.55%. Found: C, 62.14; H, 4.39; N, 15.29%.

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