

Synthesis and Biological Evaluation of Some Novel [4-(1H-Benzimidazol-2-yl)-thiazol-2-yl]-benzylidene-amines and N-[4-(1H-Benzimidazol-2-yl)-thiazol-2-yl]-N'-benzylidene-hydrazines

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ABSTRACT. A new family of thiazole heterocycles, namely [4-(1H-benzimidazol-2-yl)-thiazol-2-yl]-benzylidene-amines has been synthesized by the condensation of 4-(1H-Benzimidazol-2-yl)-thiazol-2-ylamine with various aromatic aldehydes and N-[4-(1H-benzimidazol-2-yl)-thiazol-2-yl]-N'-benzylidene-hydrazines through the cyclization of 1-(1H-benzimidazol-2-yl)-2-bromo-ethanone with arylthiosemicarbazones. The target compounds are achieved by using 1-(1H-Benzimidazol-2-yl)-ethanone as starting material. The chemical structures of all newly synthesized compounds were confirmed by their IR, ¹H NMR and Mass spectral data. Further the compounds were used to evaluate their antimicrobial activity and found that the appreciable antimicrobial activity by some of the title compounds.

Key words: Benzimidazol-2-yl-thiazole, Antibacterial activity, Antifungal activity

INTRODUCTION

Thiazole derivatives are an important class of heterocyclic compounds. They occupy an important position in medicinal chemistry, presenting a wide range of bioactivities. As medicines, many of them display including antibacterial and antifungal,^{1–3} anti-HIV,^{4,5} hypertension,^{6,7} anti-inflammatory,⁸ anticancer,⁹ anti-convulsant,¹⁰ antiinflammation,¹¹ antidepressant and tubercular activities.¹² Thiazoles and their derivatives have attracted continuing interest over the years because of their varied biological activities. Thiazoles have been incorporated into a wide variety of therapeutically interesting candidates.

Benzimidazoles are among the important heterocyclic compounds found in several natural and non-natural products such as Vitamin B₁₂,¹³ marine alkaloid kealiquinone,¹⁴ benzimidazole nucleosides.¹⁵ Some of their derivatives are marketed as anti-fungal agents such as Carbendazim,¹⁶ anti-helmintic agents such as Mebendazole and thiabendazole¹⁷ and anti-psychotic drug such as Pimozide.¹⁸ Resistance to number of anti-microbial agents among a variety of clinically significant species of bacteria is becoming increasingly important global problem. Benzimidazole ring displays an important heterocyclic pharmacophor in drug discovery. Benzimidazole derivatives are of wide interest because of their diverse biological activity and clinical applications. The compounds carrying different substituents in

the benzimidazole structure are associated with a wide range of interesting biological activities such as anti-tubercular,¹⁹ anti-cancer²⁰ and anti-coagulant properties.²¹ Further, synthesis and evaluation of different substituted benzimidazole derivatives resulted in the discovery of omeprazole, lansoprazole, rabeprazole, and pantoprazole.²² Looking at the importance of benzimidazole nucleus, it was thought that it would be worthwhile to design and synthesize some new benzimidazole derivatives and screen them for potential biological activities.

Following the successful introduction, inspired by the biological profile of thiazole and benzimidazole and their increasing importance in pharmaceutical and biological fields, and in continuation of our research on biologically active heterocycles²³ considering the scope to introduce thiazolyl moiety into the benzimidazole ring, it was thought worthwhile to undertake the synthesis of the title compounds with the view to obtain certain new chemical entities with both active pharmacophores in a single molecular framework for the potential intensified biological activities.

RESULTS AND DISCUSSION

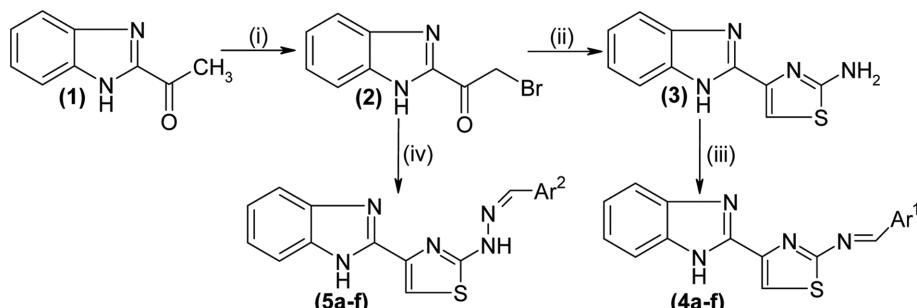
In this article, we wish to report the synthesis of novel [4-(1H-benzimidazol-2-yl)-thiazol-2-yl]-benzylidene-amines **4a–f** in good yields, from 4-(1H-benzimidazol-2-yl)-thiazol-2-ylamine **3**, which in turn is synthesized by the

reaction of 1-(1*H*-benzoimidazol-2-yl)-2-bromo-ethanone **2** with thiourea, and also the synthesis of novel *N*-[4-(1*H*-benzoimidazol-2-yl)-thiazol-2-yl]-*N'*-benzylidene hydra-zines **5a-f** in good yields, from compound 1-(1*H*-benzoimidazol-2-yl)-2-bromo-ethanone **2** and aryl-thiosemicarbazone (*Schemes 1*). The key intermediate **2**, required for the synthesis of the title compounds was prepared according to the procedure outlined in the *Scheme 1*. Acyl bromination of 1-(1*H*-benzoimidazol-2-yl)-ethanone **1** with molecular bromine in the presence of chloroform gave 1-(1*H*-benzoimidazol-2-yl)-2-bromo-ethanone **2** in good yield. Compound **2** was then on cyclocondensation with thiourea in refluxing isopropanol gave 4-(1*H*-benzoimidazol-2-yl)-thiazol-2-ylamine **3**. Subsequently condensation of compound **3** with various aromatic aldehydes in refluxing ethanol gave the title compounds [4-(1*H*-benzoimidazol-2-yl)-thiazol-2-yl]-benzylidene-amines **4a-f** in good to excel-

lent yields. Further, the intermediate **2** was treated with the various arylthiosemicarbazone (prepared by the reaction of aromatic aldehydes with thiosemicarbazide in presence of sodium acetate in acetic acid)²⁴ in refluxing isopropyl alcohol to give the other title compounds *N*-[4-(1*H*-benzoimidazol-2-yl)-thiazol-2-yl]-*N'*-benzylidene-hydrazines **5a-f** in good yields. The structures of all the newly synthesized compounds were confirmed by their IR, ¹H NMR and Mass spectral data and further the compounds **4a-f** and **5a-f** were screened for their antibacterial and antifungal activities.

ANTIMICROBIAL ACTIVITY

The activity was determined using disc diffusion method²⁵ by measuring zone of inhibition in mm. All the compounds, **4a-f** and **5a-f**, were screened in vitro at concentration of



Scheme 1. Synthesis of [4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-yl]-benzylidene-amines (**4a-f**) and *N*-[4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-yl]-*N'*-benzylidene-hydrazines (**5a-f**). (i) Br₂, CHCl₃, 0–5 °C, 3 h; (ii) NH₂CSNH₂, Isopropanol, Pyridine, Reflux, 6 h; (iii) Ar¹-CHO, Ethanol, Reflux, 2–3 h; (iv) NH₂CSNHN = CHAR², Isopropanol, Reflux, 1–2 h. Ar¹/Ar² a = C₆H₅, b = 4-NO₂-C₆H₄, c = 4-Br-C₆H₄, d = 4-CH₃O-C₆H₄, e = 3,4-(O-CH₂-O)C₆H₃, f = 2-furyl

Table 1. Antimicrobial activity of [4-(1*H*-benzoimidazol-2-yl)-thiazol-2-yl]-benzylidene-amines (**4a-f**) and *N*-[4-(1*H*-benzoimidazol-2-yl)-thiazol-2-yl]-*N'*-benzylidene-hydrazines (**5a-f**) (Zone of inhibition in mm)

Compound	Antibacterial activity					Antifungal activity	
	<i>S. aureus</i>	<i>B. subtilis</i>	<i>E. coli</i>	<i>P. aeruginosa</i>	<i>C. albicans</i>	<i>A. niger</i>	
4a	13	20	11	18	—	11	
4b	19	18	13	16	11	10	
4c	29	28	15	19	08	10	
4d	20	21	12	17	—	12	
4e	27	27	13	15	11	10	
4f	22	24	14	18	12	11	
5a	12	13	11	—	10	12	
5b	9	13	10	11	13	11	
5c	21	22	13	14	12	12	
5d	19	21	17	17	11	10	
5e	19	20	11	14	10	11	
5f	10	14	11	12	11	12	
Ciprofloxacin	26	26	28	25	—	—	
Fluconazole	—	—	—	—	26	25	

—Indicates bacteria are resistant to the compound.

5 µg/disc for their antibacterial activity against two Gram-positive strains (*Staphylococcus aureus* and *Bacillus subtilis*) and two Gram-negative strains (*Escherichia coli* and *Pseudomonas aeruginosa*). Antifungal evaluation was carried out against *Candida albicans* and *Aspergillus niger* at concentration of 5 µg/disc. Standard antibacterial drug Ciprofloxacin (5 µg/disc) and antifungal drug Fluconazole (5 µg/disc) were also tested under similar conditions against these organisms. Each experiment was done in triplicate and the average reading was taken. The results of antimicrobial studies of newly synthesized compounds reveal that the compounds possess significant antibacterial and moderate antifungal activities. Compounds **4c** and **4e** with bromo and 1,3-dioxo methylene substitution were found to be most potent compounds of the series with antibacterial activity higher than that of standard drug i.e., ciprofloxacin against *S. aureus* and *B. subtilis*. Compounds **4b**, **4f**, **5c**, **5d** and **5e** showed moderate activities against *S. aureus* and *B. subtilis*. All the other newly synthesized compounds showed either moderate activity or no activity against bacterial strains. In general, compounds **4a-f** has depicted more potent activities than compounds **5a-f**. Even though, the synthesized compounds did not exhibit appreciable antifungal activities, yet compounds **4b**, **4c**, **4e**, **4f**, **5c**, **5d** and **5e** can be chosen for further studies aimed at producing antimicrobial agents with enhanced activity. The outstanding properties of this new class of antibacterial substances deserve further investigation in order to clarify the mode of action at molecular level, responsible for the activity observed. More extensive study is also warranted to determine additional physicochemical and biological parameters to have a deeper insight into structure-activity relationship and to optimize the effectiveness of this series of molecules.

EXPERIMENTAL

Research chemicals were purchased from either Aldrich Company or Fluka and used without further purification, or were prepared according to the procedure described in the literature. Reactions were monitored by thin layer chromatography (TLC) on silica gel plates (60 F₂₅₄; Merck) visualizing with ultraviolet light or iodine vapors. The yields of the products reported here are unoptimized. Melting points were determined on a Fisher-Johns apparatus and are uncorrected. IR spectra were recorded on a Perkin-Elmer FTIR 5000 spectrometer, using KBr pellets. ¹H NMR spectra were recorded on a Varian Gemini spectrometer, operating at 300 MHz. Chemical shifts (δ)

are reported in parts per million down field from tetramethylsilane. Mass spectra were obtained on a VG micro mass 7070H spectrometer operating at 70 eV.

General Procedure for the Synthesis of 1-(1*H*-benzoimidazol-2-yl)-2-bromo-ethanone (2)

To a stirred solution of 1-(1*H*-benzoimidazol-2-yl)-ethanone (**1**) (1 mmol) in chloroform (20 ml), was added drop wise a solution of bromine (2 mmol) dissolved in chloroform (20 ml) at 0–5 °C. The mixture was further stirred for 3 h. After completion of the reaction (monitored by TLC), the resultant mixture was then poured into ice cold water. Crude product was collected by filtration, washed with water, dried, and recrystallized from ethanol to give 1-(1*H*-benzoimidazol-2-yl)-2-bromo-ethanone (**2**) in pure form.

General Procedure for the Synthesis of 4-(1*H*-benzoimidazol-2-yl)-thiazol-2-ylamine (3)

To a stirred solution of thiourea (1 mmol) in isopropanol (20 ml), was added drop wise a solution of compound (**2**) (1 mmol) in isopropanol (20 ml) over a period of 30 min. The mixture was refluxed for 3 h, and then pyridine (5 ml) was added and continued reflux for 6 h. After completion of the reaction, (monitored by TLC), the solvent was removed in vacuo. The crude product was dried and crystallized from ethanol to give pure 4-(1*H*-benzoimidazol-2-yl)-thiazol-2-ylamine (**3**).

General Procedure for the Synthesis of [4-(1*H*-benzoimidazol-2-yl)-thiazol-2-yl]-benzylidene-amines (4a-f)

A mixture of suitable aromatic aldehyde (1 mmol) and compound (**3**) (1 mmol) in ethanol (20 ml) was refluxed for 2–3 h. After completion of the reaction (monitored by TLC), the mixture was cooled and the solvent evaporated. The formed crude product was washed with cold aq. ethanol and then the product was purified by recrystallization from ethanol to afford the corresponding pure [4-(1*H*-benzoimidazol-2-yl)-thiazol-2-yl]-benzylidene-neamines (**4a-f**).

General Procedure for the Synthesis of *N*-[4-(1*H*-benzoimidazol-2-yl)-thiazol-2-yl]-*N'*-benzylidene-hydrazines (5a-f)

A mixture of suitable arylthiosemicarbazone (1 mmol) and compound (**2**) (1 mmol) in isopropanol (20 ml) was refluxed for 1–2 h. When the foaming product was formed, the mixture was allowed to cool and the solid filtered, purified by recrystallization from aq. ethanol afforded corresponding *N*-[4-(1*H*-benzoimidazol-2-yl)-thiazol-2-yl]-*N'*-benzylidene-hydrazine (**5a-f**) in pure form.

Physical and Spectral Data of Synthesized Compounds

1-(1*H*-Benzoimidazol-2-yl)-2-bromo-ethanone (2)

Yellow solid, yield: 68%; m.p: 125–127 °C; IR (KBr): cm^{-1} , 3212, 3045, 1728, 1680, 1640, 1587, 1180; ^1H NMR (300 MHz, CDCl_3): δ 4.21 (s, 2H); 7.25–7.93 (m, 4H); 13.21 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ 191.5, 139.4, 136.5, 133.2, 124.7, 123.6, 118.0, 114.2, 38.6; MS: m/z (M^+) 239.

4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-ylamine (3)

Pale yellow solid, yield: 72%; m.p: 136–138 °C; IR (KBr): cm^{-1} , 3285, 3245, 3065, 1685, 1654, 1590; ^1H NMR (300 MHz, CDCl_3): δ 7.18–7.85 (m, 4H); 7.36 (bs, 2H); 7.59 (s, 1H); 13.15 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ 168.7, 144.3, 137.2, 135.2, 133.3, 126.4, 119.6, 114.2, 111.0, 106.4; MS: m/z (M^+) 216.

[4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-yl]-benzylidene-amine (4a)

White solid, yield: 74%; m.p: 118–120 °C; IR (KBr): cm^{-1} , 3215, 3058, 1625, 1585, 1475, 1174, 640; ^1H NMR (300 MHz, CDCl_3): δ 7.17–7.86 (m, 5H); 7.28–7.92 (m, 4H); 7.48 (s, 1H); 8.21 (s, 1H); 13.21 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ 166.1, 157.4, 147.3, 143.2, 139.4, 136.2, 133.1, 131.5, 128.5, 126.5, 124.6, 121.0, 119.6, 117.3, 115.3, 112.7, 110.5; MS: m/z (M^+) 304.

[4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-yl]-[4-nitro-benzylidene]-amine (4b)

Pale yellow solid, yield: 71%; m.p: 131–133 °C; IR (KBr): cm^{-1} , 3225, 3062, 1625, 1584, 1475; ^1H NMR (300 MHz, CDCl_3): δ 7.28–7.92 (m, 4H); 7.48 (s, 1H); 7.78 (d, 2H, $J = 8.5$ Hz); 7.98 (s, 1H); 8.21 (d, 2H, $J = 8.5$ Hz); 13.21 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ 163.5, 150.2, 146.3, 145.6, 142.0, 139.7, 136.4, 134.2, 127.6, 126.5, 124.6, 123.5, 121.3, 120.5, 117.0, 113.2, 109.6; MS: m/z (M^+) 349.

[4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-yl]-[4-bromo-benzylidene]-amine (4c)

Yellow solid, yield: 70%; m.p: 141–143 °C; IR (KBr): cm^{-1} , 3432, 3058, 1618, 1574, 625, 584; ^1H NMR (300 MHz, CDCl_3): δ 7.31–7.84 (m, 4H); 7.36 (s, 1H); 7.64 (d, 2H, $J = 8.2$ Hz); 7.84 (s, 1H); 8.06 (d, 2H, $J = 8.2$ Hz); 13.06 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ 161.2, 151.3, 148.4, 144.2, 143.1, 138.4, 134.8, 132.3, 128.1, 125.2, 123.7, 122.7, 120.5, 119.2, 116.3, 114.1, 106.3; MS: m/z (M^+) 383.

[4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-yl]-[4-methoxy-benzylidene]-amine (4d)

Yellow solid, yield: 69%; m.p: 135–137 °C; IR (KBr): cm^{-1} , 3226, 3045, 1622, 1575, 1465, 645; ^1H NMR (300 MHz, CDCl_3): δ 3.72 (s, 3H); 7.06–7.82 (m, 4H); 7.36 (s, 1H); 7.74 (d, 2H, $J = 8.0$ Hz); 7.86 (s, 1H); 8.14 (d, 2H, $J = 8.0$ Hz); 13.09 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ

166.3, 159.4, 147.5, 143.2, 141.5, 136.1, 135.2, 129.7, 126.4, 124.2, 121.4, 120.5, 119.6, 117.4, 115.3, 112.3, 104.2, 54.3; MS: m/z (M^+) 334.

Benzo[1,3]dioxol-5-ylmethylene-[4-(1*H*-benzoimidazol-2-yl)-thiazol-2-yl]-amine (4e)

White solid, yield: 72%; m.p: 110–112 °C; IR (KBr): cm^{-1} , 3224, 3058, 1625, 1574, 632; ^1H NMR (300 MHz, CDCl_3): δ 5.81 (s, 2H); 6.80 (d, 1H, $J = 7.5$ Hz); 6.87 (d, 1H, $J = 7.5$ Hz); 7.21 (s, 1H), 7.15–7.90 (m, 4H); 7.62 (s, 1H); 7.81 (s, 1H); 13.12 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ 161.7, 155.2, 149.3, 146.4, 143.7, 139.5, 137.0, 131.5, 129.6, 126.4, 125.3, 123.8, 122.3, 121.5, 118.6, 116.1, 101.7, 93.6; MS: m/z (M^+) 348.

[4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-yl]-furan-2-yl-methylene-amine (4f)

Yellow solid, yield: 70%; m.p: 144–146 °C; IR (KBr): cm^{-1} , 3212, 3052, 1628, 1574; ^1H NMR (300 MHz, CDCl_3): δ 7.15–7.86 (m, 4H); 7.19–7.52 (m, 3H); 7.56 (s, 1H); 7.85 (s, 1H); 12.98 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ 165.3, 158.4, 152.3, 147.2, 144.3, 141.0, 139.5, 137.2, 134.6, 131.0, 129.6, 127.2, 126.7, 124.3, 123.0, 121.4, 119.6, 116.7, 114.3, 113.0, 108.6; MS: m/z (M^+) 294.

N-[4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-yl]-N'-benzylidene-hydrazine (5a)

Orange solid, yield: 74%; m.p: 152–154 °C; IR (KBr): cm^{-1} , 3412, 3054, 1624, 1575; ^1H NMR (300 MHz, CDCl_3): δ 7.21–7.74 (m, 5H); 7.18–7.84 (m, 4H); 7.52 (s, 1H); 8.18 (s, 1H); 9.42 (s, 1H); 13.10 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ 169.3, 157.6, 143.2, 138.6, 136.2, 135.8, 131.6, 130.2, 129.6, 128.4, 127.3, 126.4, 125.3, 124.3, 117.5, 114.3, 110.3; MS: m/z (M^+) 319.

N-[4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-yl]-N'-(4-nitro-benzylidene)-hydrazine (5b)

Yellow solid, yield: 75%; m.p: 128–130 °C; IR (KBr): cm^{-1} , 3425, 3048, 1624, 1574, 1485; ^1H NMR (300 MHz, CDCl_3): δ 7.18–7.84 (m, 4H); 7.32 (s, 1H); 7.69 (d, 2H, $J = 7.8$ Hz); 7.84 (s, 1H); 8.19 (d, 2H, $J = 7.8$ Hz); 9.49 (s, 1H); 13.15 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ 167.6, 159.3, 154.3, 141.3, 139.3, 136.2, 134.5, 133.4, 131.7, 129.7, 126.1, 123.7, 122.5, 121.7, 116.3, 112.7, 108.6; MS: m/z (M^+) 364.

N-[4-(1*H*-Benzoimidazol-2-yl)-thiazol-2-yl]-N'-(4-bromo-benzylidene)-hydrazine (5c)

Pale yellow solid, yield: 77%; m.p: 160–162 °C; IR (KBr): cm^{-1} , 3416, 3058, 1625, 1585; ^1H NMR (300 MHz, CDCl_3): δ 7.25–7.74 (m, 4H); 7.32 (s, 1H); 7.59 (d, 2H, $J = 8.0$ Hz); 7.79 (s, 1H); 8.12 (d, 2H, $J = 8.0$ Hz); 9.39 (s, 1H); 13.15 (bs, 1H); ^{13}C NMR (100 MHz, CDCl_3): δ 168.2, 156.3, 141.0, 136.8, 135.8, 134.7, 133.6, 132.3, 130.7,

126.7, 125.8, 124.8, 123.4, 122.7, 116.3, 115.4, 106.8; MS: m/z (M⁺) 398.

N-[4-(1H-Benzimidazol-2-yl)-thiazol-2-yl]-N'-(4-methoxy-benzylidene)-hydrazine (5d)

White solid, yield: 73%; m.p: 148–150 °C; IR (KBr): cm⁻¹, 3392, 3058, 1625, 1568, 1485; ¹H NMR (300 MHz, CDCl₃): δ 3.72 (s, 3H); 7.15–7.75 (m, 4H); 7.42 (s, 1H); 7.69 (d, 2H, J = 7.6 Hz); 7.84 (s, 1H); 8.16 (d, 2H, J = 7.6 Hz); 9.46 (s, 1H); 13.08 (bs, 1H); ¹³C NMR (100 MHz, CDCl₃): δ 162.4, 156.7, 145.6, 140.5, 138.6, 136.3, 134.8, 132.5, 131.2, 129.4, 126.8, 124.7, 123.6, 122.7, 120.1, 118.3, 112.4, 52.0; MS: m/z (M⁺) 349.

N-Benzo[1,3]dioxol-5-ylmethylen-N'-[4-(1H-benzimidazol-2-yl)-thiazol-2-yl]-hydrazine (5e)

Pale yellow solid, yield: 71%; m.p: 136–138 °C; IR (KBr): cm⁻¹, 3425, 3054, 1625, 1584, 1474; ¹H NMR (300 MHz, CDCl₃): δ 5.75 (s, 2H); 6.85 (d, 1H, J = 7.5 Hz); 6.94 (d, 1H, J = 7.5 Hz); 7.06 (s, 1H); 7.21–7.85 (m, 4H); 7.74 (s, 1H); 7.92 (s, 1H); 9.44 (s, 1H); 12.98 (bs, 1H); ¹³C NMR (100 MHz, CDCl₃): δ 166.7, 159.4, 149.6, 142.0, 139.7, 136.3, 134.7, 132.3, 131.0, 129.6, 126.7, 122.4, 121.8, 120.4, 119.7, 116.3, 112.5, 86.3; MS: m/z (M⁺) 363.

N-[4-(1H-Benzimidazol-2-yl)-thiazol-2-yl]-N'-furan-2-ylmethylen-hydrazine (5f)

Yellow solid, yield: 67%; m.p: 152–154 °C; IR (KBr): cm⁻¹, 3412, 3054, 1632, 1174; ¹H NMR (300 MHz, CDCl₃): δ 7.21–7.84 (m, 4H); 7.25–7.64 (m, 3H); 7.60 (s, 1H); 7.78 (s, 1H); 9.45 (s, 1H); 12.84 (bs, 1H); ¹³C NMR (100 MHz, CDCl₃): δ 171.0, 152.3, 146.7, 144.2, 143.6, 141.5, 139.7, 136.5, 127.6, 124.8, 119.6, 116.4, 112.9, 109.3, 106.2; MS: m/z (M⁺) 309.

CONCLUSION

In conclusion, a series of novel [4-(1H-benzimidazol-2-yl)-thiazol-2-yl]-benzylidene-amines **4a-f** and novel *N*-[4-(1H-benzimidazol-2-yl)-thiazol-2-yl]-*N'*-benzylidene-hydrazines **5a-f** has been designed and synthesized. The antimicrobial activity of these compounds was evaluated against various Gram-positive, Gram-negative bacteria and fungi. Among the synthesized compounds, almost all compounds showed good activity against bacteria and moderate activity against fungi and emerged as potential molecules for further development.

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