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Two step synthesis, characterization and anti-bacterial activity of series of 3-((5-chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-1,2,4-oxadiazoles

Shabhari Prasad Suggala[a], Jagadeesh Kumar Ega*[a] & A Samba Shiva Rao[b]

[a] Department of Chemistry, Chaitanya (Deemed to be University), Himayathnagar, Hyderabad 500 075, Telangana, India
[b] Department of Applied Sciences and Humanities,Maturi Venkata Subba Rao Engineering College,Nadergul, Balapur, Hyderabad 501 510, Telangana, India
E-mail: jkjagadeeshkumare@gmail.com

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A mixture of 5-chloro-1*H*-benzo[*d*]imidazole **1**, K_2CO_3 and 2-bromoacetonitrile in DMF is stirred at 60°C temperature for 8 h. to afford compound **2**. A mixture of 2-(5-chloro-1*H*-benzo[*d*]imidazol-1-yl) acetonitrile, $NH_2OH.HCl$ and triethylamine in dry DCM is stirred at room temperature for 8 hr. The aromatic carboxylic acids and Vilsmeier reagent are added and resulting mixture stirred for further 7 h at same temperature to give the crude 1,2,4-oxadiazoles **4a-l**. All the synthesised compounds have been screened for anti-bacterial evaluation. Among them **4f, 4g**, and **4h** are established to have more efficient bacterial inhibitory action against *B. subtilis*, with MICs of 3.12, 3.12, and 1.56 µg/mL, respectively, whereas typical streptomycin MICs are 6.25 µg/mL. Compound **4h** has shown more potent activity against *S. aureus*, with MIC value of 3.12 µg/mL, whereas compound **4f** has shown equipotent activity against the *S. aureus*, with MIC value of 6.25 µg/mL. Compound **4k** has shown equipotent activity against *B. subtilis* and good activity against *S. aureus* with MIC values of 3.12 µg/mL and 6.25 µg/mL respectively.

Keywords: 1,2,4-Oxadiazole, Imidazole, MIC, Anti-bacterial evaluation, Vilsmeier reagent

One of the foremost threats to human health nowadays is resistance to medications among microbial infections[1]. Therefore, it is critical to find innovative antibacterial scaffolds and develop our understanding of how to implement chemicals that have already been researched and their analogues to treat bacterial illnesses. There are rare acknowledged mechanisms of bacterial drug resistance, each of which makes a pathogen invincible for commonly used agents[2]. Particularly, in some cases, deactivation of the antibiotic occurs through enzymatic degradation or modification of the antibiotic molecule rendering it inactive. Some bacteria employ protection, alteration or overexpression of the antibiotic target. Last decade, there has been increasing of ecologically benign reagents and conditions[3–5] particularly in solvent-free procedures[6]. Imidazole derivatives possess wide-ranging spectrum of pharmacological activities[7,8]. The interesting feature of the 1,2,4-oxadiazole moiety from a therapeutic chemistry lookout is its potential coverage of a broad spectrum of therapeutic areas, including oncology[9–11], immunology[12], neurology[13–16], infectious diseases[17–20], such versatility of the 1,2,4-oxadiazole motif, in our opinion, is due to its recognition as the hydrolytically stable bio isostere of the amide and ester bonds[21–25]. Herein, we report the synthesis and biological evaluation of 3-((5-chloro-1*H*-benzo[*d*]imidazol-1-yl) methyl)-1,2,4-oxadiazoles.

Experimental Section

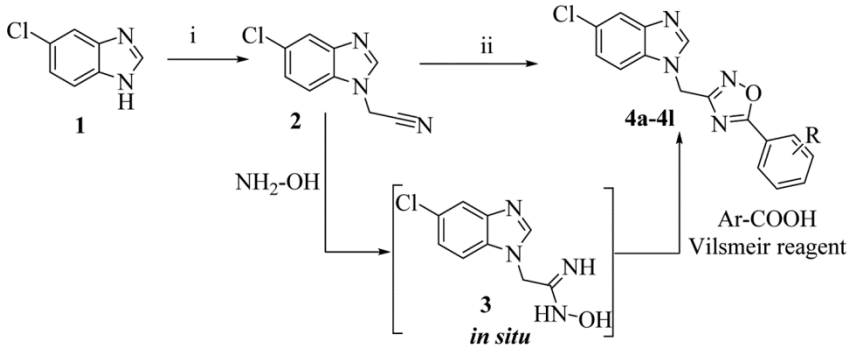
Materials and Methods

All solvents and starting materials were purchased commercially and used without additional purification. The ^1H and ^{13}C NMR spectra were acquired using a Bruker (400 MHz for ^1H and 100 MHz for ^{13}C). Mass spectra were collected using a Jeol JMC-300 spectrometer (ESI, 70 eV). The Carlo Erba 106 and PerkinElmer model 240 analyzers were used to analyse the elements. A Cintex equipment was used to determine uncorrected melting points. TLC was carried out on Merck silica gel 60 F254 precoated plates (0.25 mm), while column chromatography was carried out on silica gel (100-200 mesh). TLC with EtOAc-hexane as an eluent was employed to monitor the progress of the reactions as well as the purity of the compounds.

Results and Discussion

Initially, commercially available 5-chloro-1*H*-benzo[*d*]imidazole (**1**) was treated with 2-bromoacetonitrile using K_2CO_3 in acetone at 60°C temperature for 6 h to obtain 2-(5-chloro-1*H*-benzo[*d*]imidazol-1-yl) acetonitrile (**2**)[26,27]. This compound is confirmed by ^{1}H NMR and Mass. In ^{1}H NMR, one characteristic singlet at δ 8.09 due to the presence of C-H proton between two N-atoms in benzimidazole. A sharp singlet at δ 4.62 confirmed the presence of two protons of -CH_2 attached to the N-atom. Further, three aromatic protons were observed in the region of δ 7.89-7.63. The presence of [M+H] ion peak at *m/z* 350 in ESI-Mass spectra.

The target 1,2,4-oxadiazoles (**4a-l**) were synthesized good to excellent yields (Scheme 1, Fig. 1) by using previously well-established one-pot synthesis of intermediate 2 with NH_2-OH and different aromatic carboxylic acids using vilsmeir reagent *via* an *in situ* generated 2-(5-chloro-1*H*-benzo[*d*]imidazol-1-yl)-N-hydroxy acetimidamide (**3**) (Ref. 27). All the newly synthesized compounds (**4a-l**) were well characterized by ^{1}H NMR, ^{13}C NMR, elemental analysis, and mass (ESI-MS) spectral techniques. The ^{1}H NMR spectrum of compound **4d** showed one characteristic singlet at δ 8.12 due to the presence of imidazole proton. A sharp singlet at δ 5.33 confirmed the presence of two protons of



Reagents and Conditions: (i) 2-Bromoacetonitrile, K_2CO_3, DMF, 60°C, 8 h, (ii) (a) $NH_2OH.HCl$, TEA, DCM, RT, 8 h, (b) Vilsmeir reagents, RT, 7 h

Scheme 1 — Synthesis of 1,2,4-oxadiazoles **4a-l**

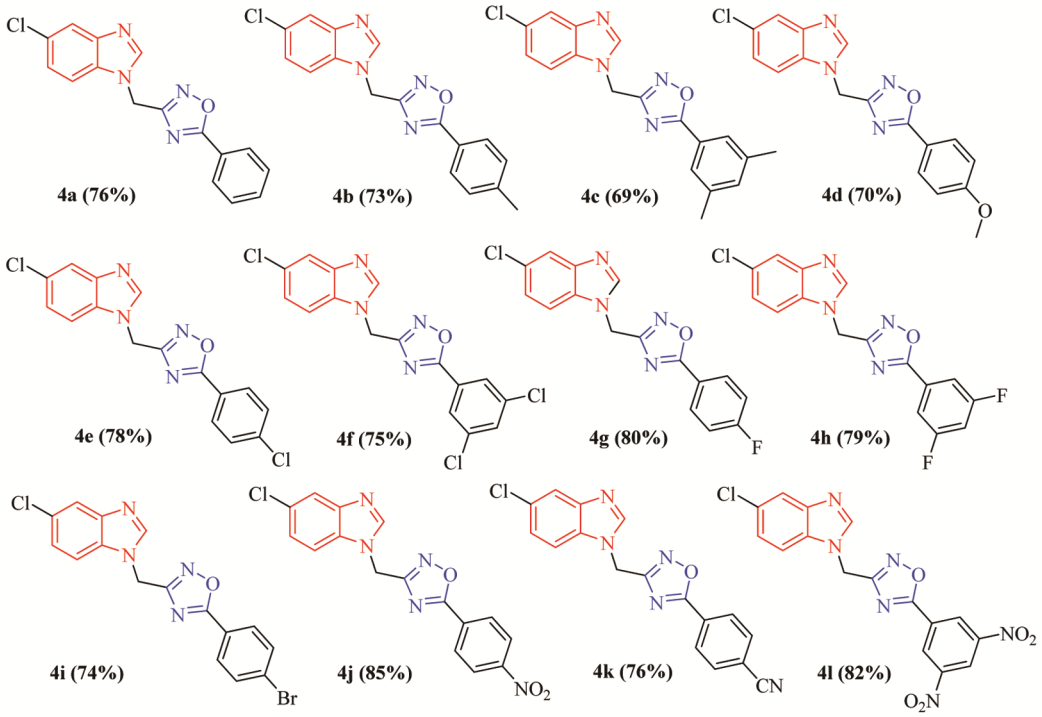


Fig. 1 — The isolated yields are given as percentages of **4a-l**

N-CH_2 attached to the oxadiazoles ring. The presence of a sharp singlet peak at δ 3.84 corresponded to the O-CH_3 protons attached to the aromatic ring. Further, all aromatic protons were observed in the region of δ 7.91-7.00. From the ^{13}C NMR, the presence of carbon signals at δ 175.28 (C-oxadiazole), δ 162.34 (C-oxadiazole), δ 160.42 (C-OCH_3), δ 56.48 (-OCH_3), and δ 42.47 (N-CH_2) confirmed the presence of characteristic carbon signals. The presence of [M+H] ion peak at *m/z* 341 in ESI-Mass spectra and the elemental analysis (CHN) data (C, 59.95; H, 3.88; N, 16.41.) confirmed molecular formula ($C_{17}H_{13}ClN_4O_2$) of compound **4d**.

Antibacterial activity

Using the conventional broth microdilution method, the title compounds (**4a-l**) were tested for *in vitro* antibacterial activity against gram-positive (G+ve) bacterial strains *B. subtilis*, *S. aureus*, and *S. epidermidis* with streptomycin serving as a positive control[28]. All derived compounds minimum inhibitory concentrations (MICs) were indicated in µg/mL. The results are shown in Table 1. Table 1 demonstrates that **4f**, **4g**, and **4h** demonstrated more efficient bacterial inhibitory action against *B. subtilis*, with MICs of 3.12, 3.12, and 1.56 µg/mL, respectively, whereas typical streptomycin MICs were 6.25 µg/mL. Compound **4h** has showed more potent activity against *S. aureus*, with MIC value of 3.12 µg/mL, whereas compound **4f** has shown equipotent activity against the *S. aureus*, with MIC value of 6.25 µg/mL. Compound **4k** shown equipotent activity against *B. subtilis* and good activity against *S. aureus* with MIC values of 3.12 µg/mL and 6.25 µg/mL, and compounds **4e** and **4i** similarly demonstrated moderate activity against *B. subtilis* and *S. aureus* (with MIC values 6.25 µg/mL). The remaining compounds were ineffective against the tested strains (Fig. 2).

Chemistry

Synthesis of 2-(5-chloro-1*H*-benzo[*d*]imidazol-1-yl) acetonitrile, 2

A mixture of 5-chloro-1*H*-benzo[*d*]imidazole (1) (5 g, 0.033 mol), K_2CO_3 (0.099 mol) and 2-bromoacetonitrile (0.038 mol) in DMF (50 mL) was stirred at 60°C temperature for 8 h. The completion of the reaction as monitored by TLC, the mixture was diluted with cold-water (50 mL) and extracted with ethyl acetate (2 × 30 mL). The combined organic layer was washed with brine (2 × 30 mL), then dried under anhydrous Na_2SO_4 and finally concentrated under vacuum to afford compound (**2**). White solid. Yield 81%.^{1}H NMR (400 MHz, DMSO-d_6): δ 8.09

Table 1 — *In vitro* antibacterial activity data of compounds **4a-l**

Compd	R	MIC (µg/mL)		
		B. subtilis	*S. aureus*	*S. epidermidis*
4a	H	50	50	–
4b	4-Me	25	25	50
4c	3,5-diMe	–	50	–
4d	4-OMe	25	50	25
4e	4-Cl	12.5	12.5	25
4f	3,5-diCl	3.12	6.25	12.5
4g	4-F	3.12	12.5	12.5
4h	3,5-diF	1.56	3.12	6.25
4i	4-Br	12.5	12.5	25
4j	4-NO_2	25	25	50
4k	4-CN	6.25	12.5	25
4l	3,5-diNO_2	25	25	50
Standard	Streptomycin	6.25	6.25	3.12

Note: "–" indicates concentration > 50 µg/mL

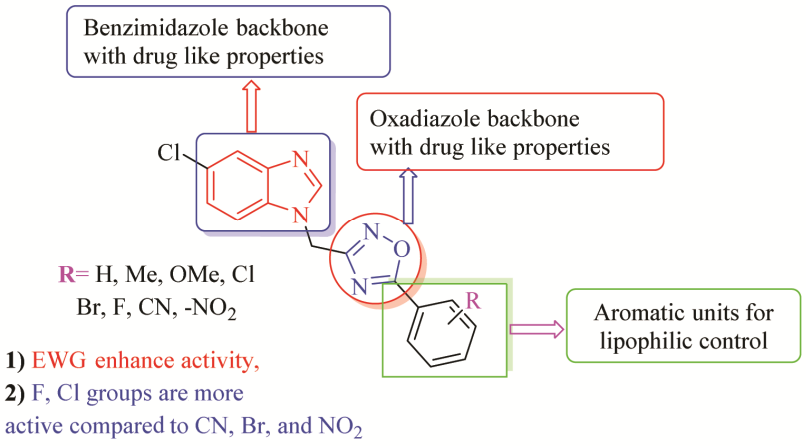


Fig. 2 — SAR of target benzimidazole based 1,2,4-oxadiazole derivatives

(s, 1H), 7.89 (s, 1H), 7.79 (d, J = 8.0 Hz, 1H), 7.63 (d, J = 8.0 Hz, 1H), 4.62 (s, 2H, N-CH_2); ESI-MS: *m/z* 192 $[M+H]^+$.

General procedure for the synthesis of fused thiochromeno[4,3-*d*]isoxazoles, 4a-l

A mixture of 2-(5-chloro-1*H*-benzo[*d*]imidazol-1-yl)acetonitrile (0.3g 1.6 mmol), $NH_2OH.HCl$ (1.7 mmol mol) and triethylamine (1 mL) in dry DCM (10 mL) was stirred at room temperature for 8 hours. After the aromatic carboxylic acids (1.7 mmol) and vilsmeier reagent (1.6 mmol) were added and resulting mixture stirred for further 7 hours at same temperature. The completion of reaction was confirmed by TLC, then the reaction mixture was washed successively with saturated $NaHCO_3$ (20 mL) and brine (20 mL). The organic layer was dried (Na_2SO_4), filtered and the solvent was removed to give the crude product, which 1,2,4-oxadiazoles (**4a-l**) were purified by short column chromatography (petroleum ether/ethyl acetate 7.5:2.5).

3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-5-phenyl-1,2,4-oxadiazole, 4a: Dirty white solid. m.p.110-112°C. ^{1}H NMR (400 MHz, DMSO-d_6) δ 8.12 (s, 1H), 7.91 (s, 1H), 7.81 (d, J = 8.0 Hz, 1H), 7.67 (d, J = 8.0 Hz, 2H), 7.62 (d, J = 8.0 Hz, 1H), 7.43-7.36 (m, 3H), 5.32 (s, 2H, N-CH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ 175.43, 162.70, 146.64, 142.72, 132.86, 131.21, 129.48, 128.09(2C), 127.75, 126.65(2C), 121.29, 119.39, 115.53, 42.64; ESI-MS: *m/z* 311 $[M+H]^+$. Anal. Calcd for $C_{16}H_{11}ClN_4O$: C, 61.84; H, 3.57; N, 18.03. Found: C, 61.87; H, 3.60; N, 18.00%.

3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-5-(p-tolyl)-1,2,4-oxadiazole, 4b: Pale red solid. m.p.116-118°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.13 (s, 1H), 7.90 (s, 1H), 7.82 (d, J = 8.0 Hz, 1H), 7.63 (d, J = 8.0 Hz, 1H), 7.59 (d, J = 8.0 Hz, 2H), 7.36 (d, J = 8.0 Hz, 2H), 5.32 (s, 2H, N-CH_2), 2.31 (s, 3H,-CH_3); ^{13}C NMR (100 MHz, DMSO-d_6) δ 175.32, 162.45, 146.61, 142.22, 140.51, 131.11, 129.65, 128.17(2C), 127.21(2C), 126.45, 121.32, 119.43, 115.41, 42.57, 21.31; ESI-MS: *m/z* 325 $[M+H]^+$. Anal. Calcd for $C_{17}H_{13}ClN_4O$: C, 62.87; H, 4.03; N, 17.25. Found: C, 62.89; H, 4.05; N, 17.21%.

3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-5-(3,5-dimethylphenyl)-1,2,4-oxadiazole, 4c: Pale red solid. m.p.121-123°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.11 (s, 1H), 7.90 (s, 1H), 7.80 (d, J = 8.0 Hz, 1H), 7.63 (d, J = 8.0 Hz, 1H), 7.52 (s, 1H), 7.21 (s, 2H), 5.31 (s, 2H, N-CH_2), 2.27 (s, 6H, 2-CH_3); ^{13}C NMR (100 MHz, DMSO-d_6): δ 175.23, 162.44, 146.67, 142.32, 138.19(2C), 131.37, 130.42, 129.18, 127.26, 124.37(2C), 121.33, 119.40, 115.51, 42.87, 20.23 (2C); ESI-MS: *m/z* 339 $[M+H]^+$. Anal. Calcd for $C_{18}H_{15}ClN_4O$: C, 63.81; H, 4.46; N, 16.54. Found: C, 63.84; H, 4.49; N, 16.51%.

3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-5-(4-methoxyphenyl)-1,2,4-oxadiazole, 4d: White solid. m.p.128-130°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.12 (s, 1H), 7.91 (s, 1H), 7.83 (d, J = 8.0 Hz, 1H), 7.70 (d, J = 8.0 Hz, 2H), 7.62 (d, J = 8.0 Hz, 1H), 7.00 (d, J = 8.0 Hz, 2H), 5.33 (s, 2H, N-CH_2), 3.84 (s, 3H,°CH_3); ^{13}C NMR (100 MHz, DMSO-d_6): δ 175.28, 162.34, 160.42, 146.18, 142.29, 131.23, 129.35, 128.06(2C), 123.68, 121.29, 119.50, 115.72, 114.88(2C), 56.48, 42.47; ESI-MS: *m/z* 341 $[M+H]^+$. Anal. Calcd for $C_{17}H_{13}ClN_4O_2$: C, 59.92; H, 3.85; N, 16.44. Found: C, 59.95; H, 3.88; N, 16.41%.

3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-5-(4-chlorophenyl)-1,2,4-oxadiazole, 4e: Pale yellow solid. m.p.125-127°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.12 (s, 1H), 7.90 (s, 1H), 7.82 (d, J = 8.0 Hz, 1H), 7.73 (d, J = 8.0 Hz, 2H), 7.63 (d, J = 8.0 Hz, 1H), 7.49 (d, J = 8.0 Hz, 2H), 5.34 (s, 2H, N-CH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ 175.76, 162.32, 146.45, 142.39, 138.12, 131.89, 130.11, 129.65(2C), 129.05, 127.66(2C), 121.34, 119.61, 115.66, 42.71; ESI-MS: *m/z* 345 $[M+H]^+$. Anal. Calcd for $C_{16}H_{10}Cl_2N_4O$: C, 55.67; H, 2.92; N, 16.23. Found: C, 55.64; H, 2.95; N, 16.25%.

3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-5-(3,5-dichlorophenyl)-1,2,4-oxadiazole, 4f: Pale yellow solid. m.p.139-131°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.13 (s, 1H), 7.93 (s, 1H), 7.82 (d, J = 8.0 Hz, 1H), 7.70 (s, 2H), 7.63 (d, J = 8.0 Hz, 1H), 7.35 (s, 1H), 5.33 (s, 2H, N-CH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ 175.34, 162.39, 146.42, 142.51, 135.42 (2C), 131.66, 130.15, 129.21, 128.09 (2C), 127.54, 121.32, 119.54, 115.65, 42.72; ESI-MS: *m/z* 379 $[M+H]^+$. Anal. Calcd for $C_{16}H_9Cl_3N_4O$: C, 50.62; H, 2.39; N, 14.76. Found: C, 50.66; H, 2.35; N, 14.73%.

3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-5-(4-fluorophenyl)-1,2,4-oxadiazole, 4g: Pale red solid. m.p.119-121°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.17 (d, J = 8.0 Hz, 2H), 8.13 (s, 1H), 7.92 (s, 1H), 7.82 (d, J = 8.0 Hz, 1H), 7.71 (d, J = 8.0 Hz,

2H), 7.63 (d, *J* = 8.0 Hz, 1H), 5.34 (s, 2H, N-CH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ 175.44, 165.21, 163.54, 162.32, 146.13, 142.65, 131.69, 130.11(2C), 128.21, 125.54, 121.30, 119.45, 116.78, 116.91, 115.44, 42.72; ESI-MS: *m/z* 329 $[M+H]^+$. Anal. Calcd for $C_{16}H_{10}ClFN_4O$: C, 58.46; H, 3.07; N, 17.04. Found: C, 58.48; H, 3.04; N, 17.01%.

3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-5-(3,5-difluorophenyl)-1,2,4-oxadiazole, 4h: Red solid. m.p.119-121°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.19 (s, 2H), 8.14 (s, 1H), 7.93 (s, 1H), 7.81 (d, *J* = 8.0 Hz, 1H), 7.75 (s, 1H), 7.63 (d, *J* = 8.0 Hz, 1H), 5.35 (s, 2H, N-CH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ 175.44, 166.31, 166.34, 164.27, 164.65, 162.37, 146.56, 142.43, 131.80, 130.32, 127.67, 122.32, 119.28, 115.78, 109.79, 109.45, 109.21, 106.88, 106.49, 106.21, 42.87; ESI-MS: *m/z* 347 $[M+H]^+$. Anal. Calcd for $C_{16}H_9ClF_2N_4O$: C, 55.43; H, 2.62; N, 16.16. Found: C, 55.40; H, 2.66; N, 16.13%.

5-(4-Bromophenyl)-3-((5-chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-1,2,4-oxadiazole, 4i: White solid. m.p.119-121°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.11 (s, 1H), 7.90 (s, 1H), 7.83 (d, *J* = 8.0 Hz, 1H), 7.66 (d, *J* = 8.0 Hz, 1H), 7.53-7.47 (m, 4H), 5.31 (s, 2H, N-CH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ 175.78, 162.32, 146.54, 142.39, 132.42(2C), 131.54(2C), 131.09, 128.21, 126.76, 124.59, 121.30, 119.52, 115.21, 42.88; ESI-MS: *m/z* 389$[M+H]^+$. Anal. Calcd for $C_{16}H_{10}BrClN_4O$: C, 49.32; H, 2.59; N, 14.38. Found: C, 49.35; H, 2.56; N, 14.35%.

3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-5-(4-nitrophenyl)-1,2,4-oxadiazole, 4j: Yellow solid. m.p.119-121°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.34 (d, *J* = 8.0 Hz, 2H), 8.14 (s, 1H), 7.92 (s, 1H), 7.86 (d, *J* = 8.0 Hz, 2H), 7.80 (d, *J* = 8.0 Hz, 1H), 7.65 (d, *J* = 8.0 Hz, 1H), 5.34 (s, 2H, N-CH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ 175.43, 162.34, 149.22, 146.54, 142.36, 131.65, 130.29, 129.76, 128.44(2C), 125.42(2C), 121.65, 119.44, 115.54, 42.76; ESI-MS: *m/z* 356$[M+H]^+$. Anal. Calcd for $C_{16}H_{10}ClN_5O_3$: C, 61.84; H, 3.57; N, 18.03. Found: C, 61.88; H, 3.53; N, 18.06%.

4-(3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-1,2,4-oxadiazol-5-yl)benzonitrile, 4k: Pale red solid. m.p.119-121°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.13 (s, 1H), 7.90 (s, 1H), 7.82 (d, *J* = 8.0 Hz, 1H), 7.71 (d, *J* = 8.0 Hz, 2H), 7.63 (d, *J* = 8.0 Hz, 1H), 7.42 (d, *J* = 8.0 Hz, 2H), 5.33 (s, 2H, N-CH_2); ^{13}C NMR (100 MHz, DMSO-d_6) δ 175.55, 162.32, 146.49, 142.86, 132.65(2C), 131.34, 130.09, 128.33, 127.65(2C), 121.43, 119.65, 119.11, 116.65, 115.87, 42.64; ESI-MS: *m/z* 336$[M+H]^+$. Anal. Calcd for $C_{17}H_{10}ClN_5O$: C, 60.82; H, 3.00; N, 20.86. Found: C, 60.85; H, 3.03; N, 20.89%.

3-((5-Chloro-1*H*-benzo[*d*]imidazol-1-yl)methyl)-5-(3,5-dinitrophenyl)-1,2,4-oxadiazole, 4l: Yellow solid. m.p.119-121°C. ^{1}H NMR (400 MHz, DMSO-d_6): δ 8.23 (s, 2H), 8.15 (s, 1H), 8.00 (s, 1H), 7.92 (s, 1H), 7.82 (d, *J* = 8.0 Hz, 1H), 7.63 (d, *J* = 8.0 Hz, 1H), 5.34 (s, 2H, N-CH_2); ^{13}C NMR (100 MHz, DMSO-d_6): δ 175.65, 162.46, 147.32(2C), 146.12, 142.64, 131.33, 129.29, 128.04(2C), 125.87, 122.45, 121.67, 119.49, 115.88, 42.64; ESI-MS: *m/z* 401$[M+H]^+$. Anal. Calcd for $C_{16}H_9ClN_6O_5$: C, 47.96; H, 2.26; N, 20.97. Found: C, 47.95; H, 2.23; N, 20.99%.

Conclusion

A series of new benzimidazole containing 1,2,4-oxadiazole derivatives (**4a–l**) were synthesized employing a one-pot synthesis of 2-(5-chloro-1*H*-benzo[*d*]imidazol-1-yl) acetonitrile, $NH_2OH.HCl$ with different aromatic carboxylic acids by using Vilsmeier reagent *via* an *in situ* generated 2-(5-chloro-1*H*-benzo[*d*]imidazol-1-yl)-N-hydroxy acetimidamides in good yields. Some of the synthesized compounds displayed moderate to excellent antibacterial activity against three gram-positive (G+ve) bacterial strains *B. subtilis*, *S. aureus*, and *S. epidermidis*. Compounds **4f**, **4g**, and **4h** showed the most effective antibacterial activity against *B. subtilis*, with MIC values ranging from 1.56–3.12 µg/mL. and also compound **4f** and **4h** also shown potent activity against S. aureus. Remaining compounds have shown moderated to poor activity against tested bacterial strains. Our further aim is the times kill studies of the human pathogenic organism using **4f** and **4h** compounds.

Supplementary Information

Supplementary information is available in the website http://nopr.niscpr.res.in/handle/123456789/58776.

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