

# Synthesis and characterization of substituted bisbenzoxazine as potent antimicrobial agent

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## ABSTRACT

Bisbenzoxazines derivatives were synthesized by refluxing substituted bisphenol and substituted aniline and formaldehyde in presence of 1, 4-dioxane with good yields. All the synthesized compounds were characterized by mass, NMR and IR and also evaluated for antimicrobial activity against four different bacterial and two fungal strains. The compounds 1c, 1h, 1j and 1l was found comparatively good active against all the bacterial strains.

**Keywords:** Benzoxazines, Synthesis, Microbial activity, Spectral techniques

## 1. INTRODUCTION

Benzoxazines are six-membered heterocyclic compounds synthesized via Mannich condensation reaction from phenols, amines, and formaldehyde. The first benzoxazine compound was synthesized by Kelly and Cone in 1944 [1]. Later, the study on benzoxazines and their oligomers was further conducted from the 1950s to 1970s [2–4]. These benzoxazines were synthesized from monophenols, monoamines, and formaldehyde, but they could not form polymers with high molecular weight. After benzoxazines based on bisphenol-A were synthesized [5, 6], a series of bifunctional benzoxazines have been prepared from bisphenols (or monophenols), monoamines (or diamines), and formaldehyde [7–15]. The bifunctional benzoxazines can form into polybenzoxazines with high molecular weight via thermally activated ring opening polymerization reaction [16–19], and the resultant polybenzoxazines possess excellent properties, such as relatively high glass transition temperature, superior mechanical properties, excellent electrical insulation properties, low moisture absorption, high thermal stability, high char yields, and low flammability. Therefore, benzoxazines can be used as high-performance matrices in electronics and aerospace industries. The work on microbial screening has not been found on bisbenzoxazine derivatives. The aim of this study is to investigate the biological activity of substituted bisbenzoxazine derivatives.

## 2. EXPERIMENTAL

All research chemicals were purchased from Allied Chemicals (Vadodara) and used as received. Thin-layer chromatography was accomplished on 0.2-mm percolated plates of silica gel G60 F254 (Merck). Visualization was made with UV light (254 and 365 nm). IR spectra were recorded on a FT-IR-8400 spectrophotometer using DRS prob.  $^1\text{H}$  (400 MHz) and  $^{13}\text{C}$  (400 MHz) NMR spectra were recorded on a Bruker AVANCE II spectrometer in  $\text{CDCl}_3$ . Chemical shifts are expressed in  $\delta$  ppm downfield from TMS as an internal standard. Mass spectra were determined using direct inlet probe on a GCMS-QP 2010 mass spectrometer (Shimadzu). Melting points were measured in open capillaries and are uncorrected.

### 2.1. General procedure for the synthesis of substituted bisbenzoxazine derivatives. (2a-l)

0.1 mol substituted aniline in 20 ml 1, 4-dioxane was slowly added to 0.2 mol formaldehyde in 80 ml, keeping temperature below  $10^\circ\text{C}$  in an ice bath. The mixture was stirred for 10 min and 0.05 mol bisphenol in 50 ml 1, 4-dioxane was added. Temperature of the mixture was raised and refluxed for 15h, cooled, poured in the crushed ice, separated yellowish solid filtered, washed well with water and dried at room temperature. Crude product was treated with 3N NaOH solution till unreacted bisphenol was removed completely by titrimetric checking with a dilute HCl solution washed well with water and dried at  $50^\circ\text{C}$ . Bisbenzoxazines were recrystallized repeatedly from appropriate solvent system to get pure product **1(a-l)**.

### 2.2 Biological testing

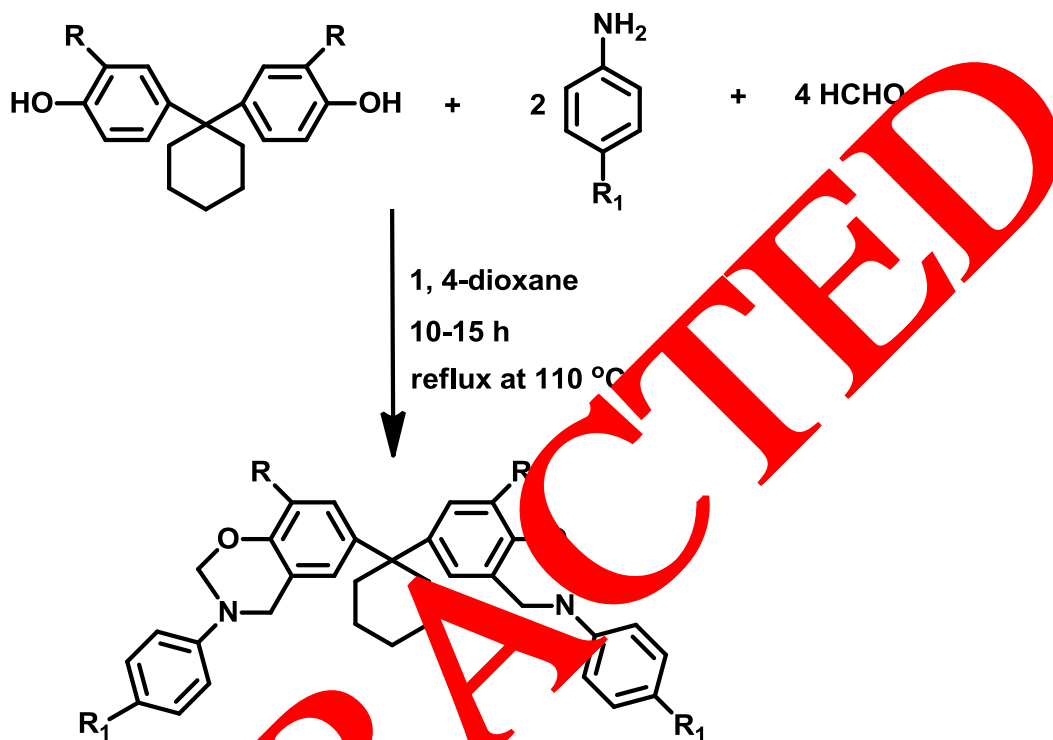
Bisphenol-C derivatives were screened for *in vitro* antibacterial and antifungal activities following micro broth dilution method [20-22]. Antibacterial activity was screened against gram-negative (*Bacillus subtilis*, *Staphylococcus aureus*) and gram-positive (*Escherichia coli*, *Salmonella typhi*) microorganisms. Antifungal activity was screened against *Aspergillus niger*, *Aspergillus clavatus* microorganisms. The standard drugs used for this study were gentamycin, ampicillin, chloramphenicol, ciprofloxacin and norfloxacin were used for antibacterial screening. Nystatin and griseofulvin were used for antifungal screening. The standard strains used for screening of antibacterial and antifungal activities were procured from Institute of Microbial Technology (IMTECH), Chandigarh, India. Mueller Hinton Broth was used as a nutrient medium for bacteria and Sabouraud Dextrose Broth for fungal growth. Inoculum size for test strain was adjusted to  $10^8$  CFU/mL by comparing the turbidity. The results were recorded in the form of primary and secondary screening. The stock solution (2000  $\mu\text{g/mL}$ ) of the compounds under investigation and standard drugs were prepared by successive dilution. In primary screening, 1000, 500 and 250  $\mu\text{g/mL}$  concentrations of the compounds were used. The compounds which found active in this primary screening were further screened. In secondary screening, 200, 100, 50, 25, 12.5 and 6.25  $\mu\text{g/mL}$  concentrations were used. The inoculated wells were incubated overnight at  $37^\circ\text{C}$  in a humid atmosphere overnight. The highest dilution showing at least 99% inhibition was considered as minimum inhibition concentration (MIC).

The investigation of antibacterial and antifungal screening data revealed that all the tested compounds 4a-t showed moderate to potent activity. The compounds 4i, 4k and 4l showed comparatively good activity against all the bacterial strains.

### 3. RESULTS AND DISCUSSION

#### Chemistry

In our current research bisbenoxazines derivatives were synthesized by refluxing substituted bisphenol-C, substituted aniline and formaldehyde in presence of 1, 4-dioxane (Scheme 1). These compounds were characterized by FTIR,  $^1\text{H}$  NMR,  $^{13}\text{C}$  NMR, and MS techniques.



Scheme 1. Synthesis of bisbenoxazine derivatives

Table 1. Substituted anilines and bisphenol-C used for the synthesis of bisbenoxazine derivatives

Code	R	R'	RT (h)	%yield	Rf values
1a	H	H	15	83	0.63(B)
1b	H	OCH <sub>3</sub>	10	78	0.74(A)
1c	H	OH	10	80	0.69(A)
1d	H	NO <sub>2</sub>	10	94	0.82(A)
1e	CH <sub>3</sub>	H	10	89	0.78(A)
1f	CH <sub>3</sub>	Cl	15	93	0.61(B)
1g	CH <sub>3</sub>	NO <sub>2</sub>	15	91	0.70(B)
1h	CH <sub>3</sub>	Br	10	84	0.64(B)
1i	Cl	H	10	87	0.64(A)
1j	Cl	OCH <sub>3</sub>	10	93	0.74(A)
1k	Br	H	10	94	0.63(A)
1l	Br	OH	10	87	0.75(A)

### 3.1. Spectroscopic data for the compounds 1(a-l)

#### 6, 6'-(cyclohexane-1,1-diyl)bis(3-phenyl)-3,4-dihydro-2H-benzo[e][1,3]oxazine) (1a):

White solid, mp 185-187°C; IR (KBr): 3459, 3327, 3193, 2999, 1648, 1586, 1261, 1061  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz, Chloroform)  $\delta$  7.33 – 7.28 (m, 2H), 7.27 – 7.09 (m, 6H), 7.08 – 6.92 (m, 2H), 6.77 – 6.66 (m, 6H), 6.02 – 5.98 (m, 2H), 5.44 – 5.40 (m, 2H), 4.85 – 4.81 (m, 2H), 4.78 – 4.74 (m, 2H), 1.82 – 1.78 (m, 2H), 1.72 (t,  $J = 9.4$  Hz, 3H), 1.46 – 1.36 (m, 5H); MS ( $m/z$ ): 502 ( $\text{M}^+$ ); Anal. Calcd for  $\text{C}_{34}\text{H}_{34}\text{N}_2\text{O}_2$ : C, 81.24; H, 6.82; N, 5.57; O, 6.37; Found: C, 81.20; H, 6.80; N, 5.52; O, 6.33.

#### 6, 6'-(cyclohexane-1,1-diyl)bis(3-(4-methoxyphenyl)-3,4-dihydro-2H-benzo[e][1,3]oxazine) (1b):

Yellow solid; mp 180-184°C; IR (KBr): 3452, 3307, 3223, 2980, 1653, 1509, 1461, 1051  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR:  $\delta$  1.37-1.44 (m, 6H, cyclohexane - $\text{CH}_2$ ), 1.70-1.78 (t, 4H, cyclohexane - $\text{CH}_2$ ), 3.83 (s, 6H, - $\text{OCH}_3$ ), 4.61 (s, 4H, - $\text{NCH}_2$ ), 6.00 (s, 4H, - $\text{OCH}_2$ ), 6.59 (m, 4H, Ar-H), 6.79-6.81 (m, 6H, Ar-H), 7.00-7.06 (m, 4H, Ar-H); MS ( $m/z$ ): 562 ( $\text{M}^+$ ); Anal. Calcd for  $\text{C}_{36}\text{H}_{38}\text{N}_2\text{O}_4$ : C, 76.84; H, 6.81; N, 4.98; O, 11.37; Found: C, 76.80; H, 6.83; N, 4.96; O, 11.33.

#### 6,6'-(cyclohexane-1,1-diyl)bis(3-(4-hydroxyphenyl)-3,4-dihydro-2H-benzo[e][1,3]oxazine) (1b):

Yellow solid; mp 210-212°C; IR (KBr): 3412, 3317, 3253, 2950, 1613, 15039, 1431, 1041  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR:  $\delta$  1.37-1.44 (m, 6H, cyclohexane - $\text{CH}_2$ ), 1.70-1.78 (t, 4H, cyclohexane - $\text{CH}_2$ ), 3.83 (s, 6H, - $\text{OCH}_3$ ), 4.61 (s, 4H, - $\text{NCH}_2$ ), 5.35 (s, 2H, -OH), 6.00 (s, 4H, - $\text{OCH}_2$ ), 6.59 (m, 4H, Ar-H), 6.77-6.79 (m, 6H, Ar-H), 7.00-7.06 (m, 4H, Ar-H); MS ( $m/z$ ): 534 ( $\text{M}^+$ ); Anal. Calcd for  $\text{C}_{34}\text{H}_{34}\text{N}_2\text{O}_4$ : C, 76.38; H, 6.41; N, 5.24; O, 11.97; Found: C, 76.34; H, 6.41; N, 5.24; O, 11.95.

#### 6, 6'-(cyclohexane-1,1-diyl)bis(3-(4-nitrophenyl)-3,4-dihydro-2H-benzo[e][1,3]oxazine) (1d)

Yellow solid; mp 194-196°C; IR (KBr): 3412, 3317, 3253, 2950, 1613, 1539, 1431, 1061  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR:  $\delta$  1.37-1.44 (m, 6H, cyclohexane - $\text{CH}_2$ ), 1.70-1.78 (t, 4H, cyclohexane - $\text{CH}_2$ ), 3.83 (s, 6H, - $\text{OCH}_3$ ), 4.61 (s, 4H, - $\text{NCH}_2$ ), 6.00 (s, 4H, - $\text{OCH}_2$ ), 6.79 (d, 2H, Ar-H), 7.00-7.06 (m, 8H, Ar-H), 8.13 (m, 4H, Ar-H); MS ( $m/z$ ): 592 ( $\text{M}^+$ ); Anal. Calcd for  $\text{C}_{34}\text{H}_{32}\text{N}_4\text{O}_6$ : C, 68.94; H, 5.44; N, 9.40; O, 16.20; Found: C, 68.88; H, 5.40; N, 9.40; O, 16.20.

#### 6,6'-(cyclohexane-1,1-diyl)bis(3-phenyl-7-methyl)-3,4-dihydro-2H-benzo[e][1,3]oxazine) (1e)

white solid; mp 194-196°C; IR (KBr): 3449, 3331, 3182, 3055, 2952, 1651, 1568, 1491, 1247, 1049  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR:  $\delta$  1.37-1.44 (m, 6H, cyclohexane - $\text{CH}_2$ ), 1.70-1.78 (t, 4H, cyclohexane - $\text{CH}_2$ ), 2.34 (s, 6H,  $\text{CH}_3$ ), 4.61 (s, 4H, - $\text{NCH}_2$ ), 5.35 (s, 2H, -OH), 6.00 (s, 4H, - $\text{OCH}_2$ ), 6.79-6.81 (m, 4H, Ar-H), 6.94 (m, 6H, Ar-H), 7.27 (m, 4H, Ar-H); MS ( $m/z$ ): 530 ( $\text{M}^+$ ); Anal. Calcd for  $\text{C}_{36}\text{H}_{38}\text{N}_2\text{O}_2$ : C, 81.47; H, 7.22; N, 5.28; O, 6.03; Found: C, 81.40; H, 7.20; N, 5.25; O, 6.00.

**6,6'-(cyclohexane-1,1-diyl)bis(3-(4-chlorophenyl-7-methyl)-3,4-dihydro-2H-benzo[e][1,3]oxazine)(1f)**

White solid; mp 245-247°C; IR (KBr): 3442, 3226, 3143, 2986, 1642, 1566, 1241, 1061  $\text{cm}^{-1}$ ;  $^1\text{H NMR}$ :  $\delta$  1.37-1.44 (m, 6H, cyclohexane - $\text{CH}_2$ ), 1.70-1.78 (t, 4H, cyclohexane - $\text{CH}_2$ ), 2.34 (s, 6H,  $\text{CH}_3$ ), 4.61 (s, 4H, - $\text{NCH}_2$ ), 6.00 (s, 4H, - $\text{OCH}_2$ ), 6.70 (d, 4H, Ar-H), 6.81 (s, 2H, Ar-H), 6.94 (s, 2H, Ar-H), 7.31 (d, 4H, Ar-H); MS ( $m/z$ ): 599 ( $\text{M}^+$ ); Anal. Calcd for  $\text{C}_{36}\text{H}_{36}\text{Cl}_2\text{N}_2\text{O}_2$ : C, 72.11; H, 6.05; Cl, 11.83; N, 4.67; O, 5.34; Found: C, 72.09; H, 6.00; Cl, 11.80; N, 4.63; O, 5.30

**6,6'-(cyclohexane-1,1-diyl)bis(3-(4-nitrophenyl-7-methyl)-3,4-dihydro-2H-benzo[e][1,3]oxazine) (1g)**

White solid; mp 165-167°C; IR (KBr): 3462, 3327, 3220, 2980, 1623, 1509, 1461, 1051  $\text{cm}^{-1}$ ;  $^1\text{H NMR}$ :  $\delta$  1.37-1.44 (m, 6H, cyclohexane - $\text{CH}_2$ ), 1.70-1.78 (t, 4H, cyclohexane - $\text{CH}_2$ ), 2.34 (s, 6H,  $\text{CH}_3$ ), 4.61 (s, 4H, - $\text{NCH}_2$ ), 6.00 (s, 4H, - $\text{OCH}_2$ ), 6.81 (s, 2H, Ar-H), 6.94 (s, 2H, Ar-H), 7.02 (d, 4H, Ar-H), 8.08 (d, 4H, Ar-H); MS ( $m/z$ ): 620 ( $\text{M}^+$ ); Anal. Calcd for  $\text{C}_{36}\text{H}_{36}\text{N}_4\text{O}_6$ : C, 69.66; H, 5.85; N, 9.03; O, 15.47; Found: C, 69.60; H, 5.82; N, 9.00; O, 15.45

**6,6'-(cyclohexane-1,1-diyl)bis(3-(4-bromophenyl-7-methyl)-3,4-dihydro-2H-benzo[e][1,3]oxazine) (1h)**

White solid; mp 256-258°C; IR (KBr): 3459, 3252, 3143, 2919, 1648, 1586, 1241, 1051  $\text{cm}^{-1}$ ;  $^1\text{H NMR}$ :  $\delta$  1.37-1.44 (m, 6H, cyclohexane - $\text{CH}_2$ ), 1.70-1.78 (t, 4H, cyclohexane - $\text{CH}_2$ ), 2.34 (s, 6H,  $\text{CH}_3$ ), 4.61 (s, 4H, - $\text{NCH}_2$ ), 6.00 (s, 4H, - $\text{OCH}_2$ ), 6.65 (d, 4H, Ar-H), 6.81 (s, 2H, Ar-H), 6.94 (s, 2H, Ar-H), 7.42 (d, 4H, Ar-H); MS ( $m/z$ ): 688 ( $\text{M}^+$ ); Anal. Calcd for  $\text{C}_{36}\text{H}_{36}\text{Br}_2\text{N}_2\text{O}_2$ : C, 62.80; H, 5.27; Br, 23.21; N, 4.07; O, 4.65; Found: C, 62.78; H, 5.25; Br, 23.18; N, 4.03; O, 4.02

**6,6'-(cyclohexane-1,1-diyl)bis(3-(4-chlorophenyl-7-chloro)-3,4-dihydro-2H-benzo[e][1,3]oxazine) (1i)**

White solid; mp 165-167°C; IR (KBr): 3462, 3327, 3220, 2980, 1623, 1509, 1461, 1051  $\text{cm}^{-1}$ ;  $^1\text{H NMR}$ :  $\delta$  1.37-1.44 (m, 6H, cyclohexane - $\text{CH}_2$ ), 1.70-1.78 (t, 4H, cyclohexane - $\text{CH}_2$ ), 2.34 (s, 6H,  $\text{CH}_3$ ), 4.61 (s, 4H, - $\text{NCH}_2$ ), 6.00 (s, 4H, - $\text{OCH}_2$ ), 6.79 (m, 2H, Ar-H), 6.94 (m, 4H, Ar-H), 6.99-7.00 (m, 4H, Ar-H), 7.27 (m, 4H, Ar-H); MS ( $m/z$ ): 571 ( $\text{M}^+$ ); Anal. Calcd for  $\text{C}_{34}\text{H}_{34}\text{Cl}_4\text{N}_2\text{O}_2$ : C, 71.45; H, 5.64; Cl, 12.41; N, 4.90; O, 5.60; Found: C, 71.45; H, 5.64; Cl, 12.40; N, 4.85; O, 5.58

**6,6'-(cyclohexane-1,1-diyl)bis(3-(4-methoxyphenyl-7-chloro)-3,4-dihydro-2H-benzo[e][1,3]oxazine) (1j)**

White solid; mp 178-180°C; IR (KBr): 3442, 3327, 3173, 2989, 1653, 1586, 1261, 1061  $\text{cm}^{-1}$ ;  $^1\text{H NMR}$ :  $\delta$  1.37-1.44 (m, 6H, cyclohexane - $\text{CH}_2$ ), 1.70-1.78 (t, 4H, cyclohexane - $\text{CH}_2$ ), 2.34 (s, 6H,  $\text{CH}_3$ ), 4.61 (s, 4H, - $\text{NCH}_2$ ), 6.00 (s, 4H, - $\text{OCH}_2$ ), 6.79 (m, 2H, Ar-H), 6.94 (m, 4H, Ar-H), 6.99-7.00 (m, 4H, Ar-H), 7.27 (m, 4H, Ar-H); MS ( $m/z$ ): 631 ( $\text{M}^+$ ); Anal. Calcd for  $\text{C}_{36}\text{H}_{36}\text{Cl}_2\text{N}_2\text{O}_4$ : C, 68.46; H, 5.75; Cl, 11.23; N, 4.44; O, 10.13; Found: C, 68.40; H, 5.70; Cl, 11.20; N, 4.40; O, 10.09

**6,6'-(cyclohexane-1,1-diyl)bis(3-phenyl-7-bromo)-3,4-dihydro-2H-benzo[e][1,3]oxazine (1k)**

White solid; mp 240-242°C; IR (KBr): 3420, 3226, 3143, 2988, 1632, 1546, 1231, 1061 cm<sup>-1</sup>; <sup>1</sup>H NMR:  $\delta$  1.37-1.44 (m, 6H, cyclohexane -CH<sub>2</sub>), 1.70-1.78 (t, 4H, cyclohexane -CH<sub>2</sub>), 2.34 (s, 6H, CH<sub>3</sub>), 4.61 (s, 4H, -NCH<sub>2</sub>), 6.00 (s, 4H, -OCH<sub>2</sub>), 6.79 (m, 2H, Ar-H), 6.94-6.95 (m, 4H, Ar-H), 7.04 (s, 2H, Ar-H), 7.27 (m, 4H, Ar-H); MS (*m/z*): 660 (M<sup>+</sup>); Anal. Calcd for C<sub>34</sub>H<sub>32</sub>Br<sub>2</sub>N<sub>2</sub>O<sub>2</sub>: C, 61.83; H, 4.88; Br, 24.20; N, 4.24; O, 4.85; Found: C, 61.79; H, 4.84; Br, 24.15; N, 4.20; O, 4.80.

**6,6'-(cyclohexane-1,1-diyl)bis(3-(4-hydroxyphenyl)-7-bromo)-3,4-dihydro-2H-benzo[e][1,3]oxazine (1l)**

White solid; mp 194-196°C; IR (KBr): 3412, 3317, 3253, 2950, 1613, 1529, 1431, 1061 cm<sup>-1</sup>; <sup>1</sup>H NMR:  $\delta$  1.37-1.44 (m, 6H, cyclohexane -CH<sub>2</sub>), 1.70-1.78 (t, 4H, cyclohexane -CH<sub>2</sub>), 2.34 (s, 6H, CH<sub>3</sub>), 4.61 (s, 4H, -NCH<sub>2</sub>), 5.35 (s, 2H, -OH), 6.00 (s, 4H, -OCH<sub>2</sub>), 6.59 (t, 4H, Ar-H), 6.77 (d, 4H, Ar-H), 6.95 (s, 2H, Ar-H), 7.04 (s, 2H, Ar-H); MS (*m/z*): 692 (M<sup>+</sup>); Anal. Calcd for C<sub>34</sub>H<sub>32</sub>Br<sub>2</sub>N<sub>2</sub>O<sub>4</sub>: C, 58.97; H, 4.66; Br, 23.08; N, 4.05; O, 9.24; Found: C, 58.94; H, 4.60; Br, 23.08; N, 4.05; O, 9.20.

**Table 2** Microbial activity of 1-phenol-C derivatives

Compound	Antibacterial activity				Antifungal activity	
	<i>E. coli</i>	<i>S. typhi</i>	<i>S. typhi</i>	<i>S. aureus</i>	<i>A. niger</i>	<i>A. clavattus</i>
	MTCC 443	MTCC 96	MTCC 41	MTCC 96	MTCC 282	MTCC 1323
1a	125	200	200	250	1000	250
1b	200	200	250	200	>1000	500
1c	<b>62.5</b>	<b>100</b>	<b>125</b>	<b>200</b>	500	500
1d	250	250	500	500	500	>1000
1e	250	200	500	250	1000	>1000
1f	125	250	250	500	500	>1000
1g	250	250	200	250	>1000	500
1h	<b>100</b>	200	<b>62.5</b>	<b>250</b>	<b>100</b>	500
1i	125	500	100	100	250	>1000
1j	<b>62.5</b>	<b>125</b>	<b>250</b>	<b>200</b>	250	>1000
1k	<b>250</b>	<b>125</b>	<b>125</b>	<b>100</b>	>1000	>1000
1l	<b>62.5</b>	250	<b>200</b>	<b>200</b>	1000	1000
Gentamycin	0.05	5	1	0.25	-	-
Ampicillin	100	100	250	250	-	-
Chloramphenicol	50	50	50	50	-	-
Ciprofloxacin	25	25	50	50	-	-
Norfloxacin	10	10	100	10	-	-
Nystatin	-	-	-	-	100	100
Griseofulvin	-	-	-	-	100	100

Bold digits indicate that synthesized compounds are comparatively active as standard drugs DMF used as control and its antibacterial activity is nil or zero

#### 4. CONCLUSION

In summary, we have described the synthesis of substituted bisbenzoxazines derivatives in excellent yields by simple refluxing. The spectral data are incorporate with the structure of compounds **1a-1**. The antimicrobial data reported here which may be the better reference for the future research in the class of pyrimidine derivatives.

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#### References

- [1] Holly F.W, Cope A. C., *J Am Chem Soc.* 66 (1944) 1875.
- [2] Burke W.J, Weatherbee C., *J Am Chem Soc.* 72 (1950) 4691.
- [3] Burke W.J, Stephens C.W. *J Am Chem Soc.* 74 (1952) 1518.
- [4] Burke W.J, Murdoch K.C, Ec. G., *J Am Chem Soc.* 76 (1954) 1677.
- [5] Ning X, Ishida H., *J Polym Sci Part A.* 32 (1994) 1121.
- [6] Ning X, Ishida H., *J Polym Sci Part B.* 32 (1994) 921.
- [7] Liu Y.F, Yue Z.Q, Gao G., *Polymer.* 51 (2010) 3722.
- [8] Jia K, Xu M.Z, Zhao R, Liu J.B, *Polym Int.* 60 (2011) 414.
- [9] Chen K.C, H.T, Chen W.B, Liao C.H, Sun K.W, Chang F.C., *Polym Int.* 60 (2011)436.
- [10] Li S.H, Wang L., *J Appl Polym Sci.* 99 (2006) 1359.
- [11] Lin C.H, Chang S.L, Hsieh C.W, Lee H.H., *Polymer.* 49 (2008) 1220.
- [12] Spontoni M, Larrechi M.S, Ronda J.C, Galia' M, Ca'diz V., *J Polym Sci Part A.* 46 (2008) 7162.
- [13] Andronesu C, Ga'rea S.A, Deleanu C, Iovu H., *Thermochim Acta.* 530 (2012) 42.
- [14] Chozhan C.K, Alagar M, Gnanasundaram P., *Acta Mater.* 57 (2009) 782.
- [15] Chang S.L, Lin C.H. Facile., *J Polym Sci Part A.* 48 (2010) 2430.

- [16] Ishida H, Rodriguez Y., *Polymer*. 36 (1995) 3151.
- [17] Russell V.M, Koenig J.L, Low H.Y, Ishida H., *J Appl Polym Sci*. 70 (1998) 1413.
- [18] Wang YX, Ishida H., *Polymer*. 40 (1999) 4563.
- [19] Wang YX, Ishida H., *Macromolecules*. 33 (2000) 2839.
- [20] National committee for clinical laboratory standards.,5th ed.; NCCLS: Wayne, PA, 2000.
- [21] Patel, N. B.; Shaikh, A. R., *Indian J. Chem.* 49 (2010) 929.

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