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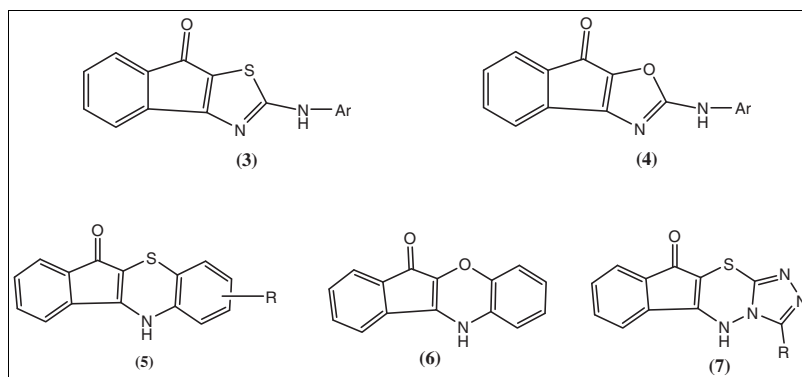
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Indandione **1** was brominated to yield 2-bromoIndandione **2**, which further reacted with substituted thiocarbamides, carbamides, 2-aminothiophenols, 2-aminophenol, and triazole to furnished 3-substituted aniline-2-thia-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene **3**, 3-substituted aniline-2-oxa-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene **4**, 2-Thia-5-aza-9-oxo-3,4-(3'-substituted)benzo-7,8-benzo-bicyclo[4.3.0]-1(6) nonene **5**, 2-oxa-5-aza-9-oxo-(3,4)-(7,8)-dibenzo-bicyclo[4.3.0]-1(6) nonene **6** and 3'-substituted-(1',2',4')triazolo[5,6-b][indeno(2,3-e)]-1,3,4-thiadiazine **7**, respectively. The structures of compounds were elucidated on the basis of spectral techniques, further the representative compounds were screened for their antimicrobial activity.

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## INTRODUCTION

Heterocyclic compounds constitute the largest family of organic compounds, regardless of structure and functionality. Heterocyclic compounds are of particular interest in medicinal chemistry and this has catalyzed the discovery and development of many new heterocyclic compounds.

In the published reports, the compounds bearing thiazole and oxazole moiety have been found to possess antibacterial [1], antitubercular [2], and anti-inflammatory [3] activity. Similarly, heterocycles containing thiazine and oxazine moiety are well-known for their diverse biological activity and play a key role as anti-psychotic [4], antiviral [5], and anti-microbial agents [6]. Fused s-triazoles and their derivatives have been investigated for their potential pharmacological properties such as antifungal [7], antidepressant [8], and plant growth regulators [9]. In view of the biological potential of the above pharmacophore, syntheses of various derivatives have been undertaken.

## RESULTS AND DISCUSSION

The synthesis of 2-bromo-Indandione **2** was achieved from Indandione **1** using bromine in glacial acetic acid.

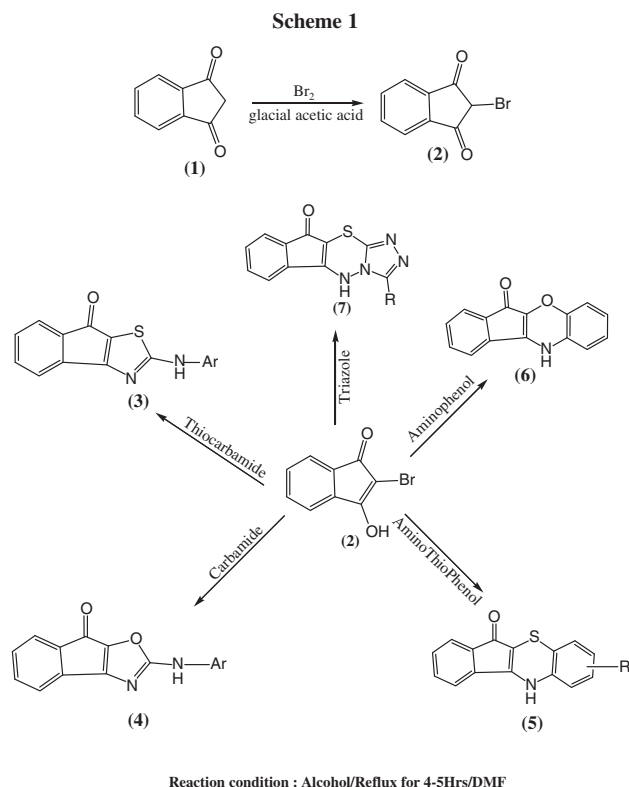
The title compounds 3-(substituted)-imino-2-thia-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene **3a-f**, 3-(substituted)-imino-2-oxa-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene **4a-f**, 2-thia-5-aza-9-oxo-3,4-(3'-substituted)benzo-7,8-benzo-bicyclo[4.3.0]-1(6) nonene **5a-c**, 2-oxa-5-aza-9-oxo-(3,4)-(7,8)-dibenzo-bicyclo[4.3.0]-1(6) nonene **6** and 3-substituted-(1,2,4)triazolo[4,5-b][indeno(2,3-e)]-1,3,4-thiadiazine **7a-c** were synthesized by reacting bromo compound **2** with substituted thiocarbamides [10], carbamides [11], 2-aminothiophenols [12, 13], 2-aminophenol and triazoles, respectively, in presence of dimethylformamide (DMF) as a catalyst and ethanol as a solvent (Scheme 1).

The structures of representative compounds were elucidated on the basis of spectral techniques, further the representative compounds were screened for their antimicrobial activity, which showed a promising activity against gram positive as well as gram negative bacterias.

The physical characterization data was given in Table 1.

## EXPERIMENTAL

All chemicals were supplied by E. Merck (Germany) and S. D. Fine Chemicals (India). Melting points of all synthesized compounds were determined in open capillary tubes using Veego



VMP-1 melting point apparatus and are expressed in degree celsius. The progress of the reaction was monitored *in situ* by a thin layer chromatography (TLC) on silica gel-coated aluminum plates as adsorbent and UV light as visualizing agent. IR spectra in KBr pellets were recorded on Perkin-Elmer spectrophotometer in the range of 4000-400  $\text{cm}^{-1}$ .  $^1\text{H}$  NMR spectra were obtained

using Varian 500 MHz NMR spectrophotometer using  $\text{CDCl}_3/\text{DMSO}-d_6$  as solvent and TMS as an internal standard. C, H, N analyses were performed on Carlo Erba 1108 (C H N) Elemental Analyzer.

**Synthesis of 2-bromo-indandione, 2.** Indandione 1 (0.01 mol) was dissolved in 10 mL of glacial acetic acid. A solution of bromine (0.01 mol) in glacial acetic acid was added dropwise with continuous stirring in presence of UV light. The stirring was continued for 1 h. The reaction mixture was quenched into ice-cold water and the product was separated out, filtered, washed with cold water. The product was purified by recrystallization from ethanol to give 2-bromo-indandione 2, yield 86%, m.p. 98–101°C.

**3-(Substituted)-imino-2-thia-4-aza-6,7-benzo-8-oxo-bicyclo [3.3.0]-1(5),3-octadiene, 3a-f.** An equimolar mixture of compound 2 (0.01 mol) and substituted thiocarbamide (0.01 mol) in ethanol (20 mL) was refluxed in presence of DMF (0.02 mol) for about 4–5 h. The progress of reaction was monitored on TLC. Upon completion of reaction, the reaction mixture was quenched into crushed ice. The product precipitated out was filtered, washed with water, and purified by recrystallization from ethanol to give thiazoles 3a-f.

**3-Imino-2-thia-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (3a).** M.p. = >260°C, Yield = 88%, IR ( $\text{cm}^{-1}$ ): 3158, 1252,  $^1\text{H}$  NMR (500 MHz,  $\text{DMSO}-d_6$ ,  $\delta$  ppm): 4.1 (s, 1H, NH), 7.15–8.18 (m, 4H, ArH), 9.79 (s, 1H, NH),  $^{13}\text{C}$  NMR (500 MHz,  $\text{DMSO}-d_6$ , ppm): 115.28, 119.45(C=C), 122.31, 124.23, 125.32, 128.21, 128.23, 131.42, 133.47, 135.56, 136.10, 138.89 (Ar C), 154.25 (C=N), 180.72 (C=O). Calcd for  $\text{C}_{10}\text{H}_6\text{ON}_2\text{S}$ : C, 59.41; H, 2.97; N, 13.86%. Found: C, 59.36; H, 2.91; N, 13.81%.

**3-(Phenyl)-imino-2-thia-4-aza-6,7-benzo-8-oxo-bicyclo [3.3.0]-1(5),3-octadiene (3b).** M.p. = 245–247°C, Yield = 89%, IR ( $\text{cm}^{-1}$ ): 3148, 1286,  $^1\text{H}$  NMR (500 MHz,  $\text{DMSO}-d_6$ ,  $\delta$  ppm): 7.18–8.28 (m, 9H, ArH), 9.68 (s, 1H, NH),  $^{13}\text{C}$  NMR (500 MHz,  $\text{DMSO}-d_6$ , ppm): 113.48, 119.24 (C=C), 122.58, 122.72, 124.36, 127.21, 129.56, 131.0, 131.31, 135.36, 136.89, 137.84 (Ar C), 157.23 (C=N), 180.77 (C=O). Calcd for  $\text{C}_{16}\text{H}_{10}\text{ON}_2\text{S}$ :

**Table 1**

Physical characterization of synthesized compounds.

Compound	Ar/R	Mol. formula	M.p. (°C)	Yields (%)
3a	H	$\text{C}_{10}\text{H}_6\text{ON}_2\text{S}$	>260	88
3b	$\text{C}_6\text{H}_5$	$\text{C}_{16}\text{H}_{10}\text{ON}_2\text{S}$	245–247	89
3c	4-Cl- $\text{C}_6\text{H}_4$	$\text{C}_{16}\text{H}_9\text{ON}_2\text{SCl}$	220–223	92
3d	4-OCH <sub>3</sub> - $\text{C}_6\text{H}_4$	$\text{C}_{17}\text{H}_{12}\text{O}_2\text{N}_2\text{S}$	210–213	87
3e	3-Cl- $\text{C}_6\text{H}_4$	$\text{C}_{16}\text{H}_9\text{ON}_2\text{SCl}$	210–213	89
3f	4-CH <sub>3</sub> - $\text{C}_6\text{H}_4$	$\text{C}_{17}\text{H}_{12}\text{ON}_2\text{S}$	228–231	87
4a	H	$\text{C}_{10}\text{H}_6\text{O}_2\text{N}_2$	178–180	89
4b	$\text{C}_6\text{H}_5$	$\text{C}_{16}\text{H}_{10}\text{O}_2\text{N}_2$	172–174	82
4c	4-Cl- $\text{C}_6\text{H}_4$	$\text{C}_{16}\text{H}_9\text{O}_2\text{N}_2\text{Cl}$	164–166	81
4d	4-OCH <sub>3</sub> - $\text{C}_6\text{H}_4$	$\text{C}_{17}\text{H}_{12}\text{O}_3\text{N}_2$	155–159	87
4e	4-NO <sub>2</sub> - $\text{C}_6\text{H}_4$	$\text{C}_{16}\text{H}_9\text{O}_4\text{N}_3$	207–208	86
4f	4-CH <sub>3</sub> - $\text{C}_6\text{H}_4$	$\text{C}_{17}\text{H}_{12}\text{O}_2\text{N}_2$	180–184	89
5a	H	$\text{C}_{15}\text{H}_9\text{NOS}$	146–148	89
5b	Cl	$\text{C}_{15}\text{H}_8\text{NOSCl}$	145–147	84
5c	CH <sub>3</sub>	$\text{C}_{16}\text{H}_{11}\text{NOS}$	120–124	82
6	H	$\text{C}_{15}\text{H}_9\text{NO}_2$	187–189	91
7a	H	$\text{C}_{11}\text{H}_6\text{N}_4\text{OS}$	215–217	81
7b	CH <sub>3</sub>	$\text{C}_{12}\text{H}_8\text{N}_4\text{OS}$	201–203	87
7c	$\text{C}_2\text{H}_5$	$\text{C}_{13}\text{H}_{10}\text{N}_4\text{OS}$	229–231	84

C, 69.06; H, 3.60; N, 10.07%. Found: C, 69.02; H, 3.52; N, 10.01%.

**3-(4'-Chlorophenyl)-imino-2-thia-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (3c).** M.p. = 220–223°C, Yield = 92%, IR (cm<sup>-1</sup>): 3368, 1705, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 7.18–8.62 (m, 8H, ArH), 9.81 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 114.22, 120.73 (C=C), 122.56, 123.41, 124.23, 125.32, 128.94, 130.45, 131.41, 135.32, 136.54, 139.95 (Ar C), 156.34 (C=N), 182.73 (C=O). Calcd for C<sub>16</sub>H<sub>9</sub>N<sub>2</sub>O<sub>2</sub>SCl: C, 61.44; H, 2.88; N, 8.96%. Found: C, 61.34; H, 2.78; N, 9.01%.

**3-(4'-Methoxyphenyl)-imino-2-thia-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (3d).** M.p. = 210–213°C, Yield = 87%, IR (cm<sup>-1</sup>): 3368, 1776, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 3.91 (s, 3H, OCH<sub>3</sub>), 7.08–8.32 (m, 8H, ArH), 9.76 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 55.46 (OCH<sub>3</sub>), 114.20, 120.73 (C=C), 121.71, 122.38, 123.42, 124.56, 125.84, 127.85, 129.65, 131.24, 138.21, 139.96 (Ar C), 154.33 (C=N), 183.74 (C=O). Calcd for C<sub>17</sub>H<sub>12</sub>N<sub>2</sub>O<sub>2</sub>S: C, 66.22; H, 3.92; N, 9.08%. Found: C, 66.32; H, 4.02; N, 9.12%.

**3-(3'-Chlorophenyl)-imino-2-thia-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (3e).** M.p. = 210–213°C, Yield = 89%, IR (cm<sup>-1</sup>): 3368, 1726, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 6.98–8.08 (m, 8H, ArH), 9.74 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 113.87, 119.47 (C=C), 122.37, 124.38, 125.49, 127.35, 128.43, 129.87, 132.38, 135.56, 136.56, 137.89 (Ar C), 158.3 (C=N), 186.7 (C=O). Calcd for C<sub>16</sub>H<sub>9</sub>N<sub>2</sub>O<sub>2</sub>SCl: C, 61.44; H, 2.88; N, 8.96%. Found: C, 61.38; H, 2.80; N, 8.89%.

**3-(4'-Methylphenyl)-imino-2-thia-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (3f).** M.p. = 228–231°C, Yield = 87%, IR (cm<sup>-1</sup>): 3338, 1726, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 2.24 (s, 3H, CH<sub>3</sub>), 6.76–7.85 (m, 8H, ArH), 9.68 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 23.35 (CH<sub>3</sub>), 112.96, 119.87 (C=C), 123.35, 124.36, 125.42, 126.73, 128.57, 130.19, 131.14, 135.36, 136.35, 137.89 (Ar C), 153.43 (C=N), 185.47 (C=O). Calcd for C<sub>17</sub>H<sub>12</sub>N<sub>2</sub>O<sub>2</sub>S: C, 69.86; H, 4.11; N, 9.59%. Found: C, 69.82; H, 4.02; N, 9.48%.

**3-(Substituted)-imino-2-oxa-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene, 4a–f.** A mixture of compound **2** (0.01 mol) and substituted carbamides (0.01 mol) in ethanol (20 mL) was refluxed in presence of DMF (0.02 mol) for about 4–5 h. The progress of reaction was monitored on TLC. Upon completion of reaction, the reaction mixture was quenched into crushed ice. The product precipitated out was filtered, washed with water and purified by recrystallization from ethanol to give oxazoles **4a–f**.

**3-Imino-2-oxa-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (4a).** M.p. = 178–180°C, Yield = 89%, IR (cm<sup>-1</sup>): 3473, 1766, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 4.28 (s, 1H, NH), 6.98–7.98 (m, 4H, ArH), 9.85 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 115.1, 119.4 (C=C), 122.4, 123.45, 125.47, 126.32, 127.43, 128.59, 130.43, 131.23, 135.52, 131.7 (Ar C), 152.6 (C=N), 179.5 (C=O). Calcd for C<sub>10</sub>H<sub>6</sub>N<sub>2</sub>O<sub>2</sub>: C, 64.52; H, 3.23; N, 15.05%. Found: C, 64.48; H, 3.17; N, 15.01%.

**3-(Phenyl)-imino-2-oxa-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (4b).** M.p. = 172–174°C, Yield = 82%, IR (cm<sup>-1</sup>): 3373, 1696, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 6.87–7.54 (m, 9H, ArH), 9.68 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 114.89, 120.40 (C=C), 121.94, 122.45, 123.57, 124.35, 126.43, 128.53, 130.75, 131.43, 132.31, 135.76 (ArC), 154.66 (C=N), 180.56 (C=O). Calcd for C<sub>16</sub>H<sub>10</sub>N<sub>2</sub>O<sub>2</sub>: C, 73.28; H, 3.81; N, 10.69%. Found: C, 73.20; H, 3.74; N, 10.62%.

**3-(4'-Chlorophenyl)-imino-2-oxa-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (4c).** M.p. = 164–166°C, Yield = 81%, IR (cm<sup>-1</sup>): 3373, 1736, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 6.78–7.86 (m, 8H, ArH), 9.72 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 113.89, 120.24 (C=C), 123.67, 124.45, 126.63, 127.43, 128.54, 129.87, 130.75, 131.43, 131.87, 132.76 (Ar C), 155.67 (C=N), 183.68 (C=O). Calcd for C<sub>16</sub>H<sub>9</sub>N<sub>2</sub>O<sub>2</sub>Cl: C, 64.76; H, 3.04; N, 9.44%. Found: C, 64.71; H, 2.94; N, 9.41%.

**3-(4'-Methoxyphenyl)-imino-2-oxa-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (4d).** M.p. = 155–159°C, Yield = 87%, IR (cm<sup>-1</sup>): 3343, 1698, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 3.84 (s, 3H, OCH<sub>3</sub>), 7.26–8.18 (m, 8H, ArH), 9.76 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 56.05 (OCH<sub>3</sub>), 114.61, 119.86 (C=C), 124.47, 125.48, 126.52, 127.45, 127.98, 129.58, 131.24, 132.45, 133.18, 134.42 (Ar C), 154.68 (C=N), 180.57 (C=O). Calcd for C<sub>17</sub>H<sub>12</sub>N<sub>2</sub>O<sub>3</sub>: C, 69.86; H, 4.11; N, 9.59%. Found: C, 69.81; H, 4.02; N, 9.51%.

**3-(4'-Nitrophenyl)-imino-2-oxa-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (4e).** M.p. = 207–208°C, Yield = 86%, IR (cm<sup>-1</sup>): 3473, 1756, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 7.25–8.28 (m, 8H, ArH), 9.69 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 114.76, 120.04 (C=C), 121.98, 122.74, 123.45, 123.87, 124.87, 125.47, 126.41, 127.65, 128.15, 129.87 (Ar C), 156.68 (C=N), 182.52 (C=O). Calcd for C<sub>16</sub>H<sub>9</sub>N<sub>3</sub>O<sub>4</sub>: C, 62.54; H, 2.93; N, 13.68%. Found: C, 62.48; H, 2.84; N, 13.61%.

**3-(4'-Methylphenyl)-imino-2-oxa-4-aza-6,7-benzo-8-oxo-bicyclo[3.3.0]-1(5),3-octadiene (4f).** M.p. = 180–184°C, Yield = 89%, IR (cm<sup>-1</sup>): 3453, 1756, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 2.22 (s, 3H, CH<sub>3</sub>), 7.28–8.14 (m, 8H, ArH), 9.74 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 23.91 (CH<sub>3</sub>), 115.16, 119.48 (C=C), 122.64, 123.84, 124.54, 125.08, 126.84, 127.24, 128.12, 129.51, 130.41, 131.52 (Ar C), 155.69 (C=N), 180.53 (C=O). Calcd for C<sub>17</sub>H<sub>12</sub>N<sub>2</sub>O<sub>2</sub>: C, 73.90; H, 4.35; N, 10.14%. Found: C, 73.84; H, 4.32; N, 10.19%.

**2-Thia-5-aza-9-oxo-3,4-(substituted)benzo-7,8-benzo-bicyclo[4.3.0]-1(6)nonene, 5a–c.** An equimolar mixture of compound **2** (0.01 mol) and substituted aminothiophenol (0.01 mol) in ethanol (20 mL) was refluxed in presence of DMF (0.02 mol) for about 4–5 h. The progress of reaction was monitored on TLC. Upon completion of reaction, the reaction mixture was quenched into crushed ice. The product precipitated out was filtered, washed with water, and purified by recrystallization from ethanol to give Thiazines **5a–c**.

**2-Thia-5-aza-9-oxo-3,4-benzo-7,8-benzo-bicyclo[4.3.0]-1(6)nonene (5a).** M.p. = 146–148°C, Yield = 89%, IR (cm<sup>-1</sup>): 3290, 1760, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 6.87–7.98 (m, 8H, ArH), 9.84 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 118.26, 120.37 (C=C), 121.82, 122.41, 123.21, 123.89, 124.51, 125.41, 126.84, 127.15, 129.85, 130.45, 132.14, 135.68 (Ar C), 179.25 (C=O). Calcd for C<sub>15</sub>H<sub>9</sub>NOS: C, 71.71; H, 3.59; N, 5.58%. Found: C, 71.64; H, 3.48; N, 5.49%.

**2-Thia-5-aza-9-oxo-3,4-(3'-chloro)benzo-7,8-benzo-bicyclo[4.3.0]-1(6)nonene (5b).** M.p. = 145–147°C, Yield = 84%, IR (cm<sup>-1</sup>): 3360, 1680, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 7.28–8.16 (m, 8H, ArH), 10.42 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 118.18, 120.63 (C=C), 123.84, 124.74, 125.21, 126.63, 127.65, 129.74, 130.21, 131.28, 132.56, 133.52, 134.08, 135.74 (Ar C), 182.84 (C=O). Calcd for C<sub>15</sub>H<sub>8</sub>NOSCl: C, 63.05; H, 2.80; N, 4.90%. Found: C, 63.01; H, 2.73; N, 4.78%.

**2-Thia-5-aza-9-oxo-3, 4-(3'-methyl) benzo-7,8-benzo-bicyclo[4.3.0]-1(6) nonene (5c).** M.p. = 120–124°C, Yield = 82%, IR (cm<sup>-1</sup>): 3370, 1690, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 2.48 (s, 3H, CH<sub>3</sub>), 7.28–8.25 (m, 8H, ArH), 10.40 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 23.91 (CH<sub>3</sub>), 118.20, 120.32 (C=C), 121.84, 122.75, 124.46, 125.62, 126.28, 128.54, 129.65, 131.24, 132.45, 133.51, 134.43, 135.62 (ArC), 180.58 (C=O). Calcd for C<sub>16</sub>H<sub>11</sub>N<sub>2</sub>O: C, 72.43; H, 4.18; N, 5.28%. Found: C, 72.34; H, 4.23; N, 5.19%.

**2-Oxa-5-aza-9-oxo-(3,4)-(7,8)-dibenzo-bicyclo[4.3.0]-1(6) nonene (6).** An equimolar mixture of compound **2** (0.01 mol) and aminophenol (0.01 mol) in ethanol (20 mL) was refluxed in presence of DMF (0.02 mol) for about 4–5 h. The progress of reaction was monitored on TLC. Upon completion of reaction, the reaction mixture was quenched into crushed ice. The product precipitated out was filtered, washed with water, and purified by recrystallization from ethanol to yield **6**.

**2-Oxa-5-aza-9-oxo-(3,4)-(7,8)-dibenzo-bicyclo[4.3.0]-1(6) nonene (6).** M.p. = 187–189°C, Yield = 91%, IR (cm<sup>-1</sup>): 3287, 1757, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 6.87–7.98 (m, 8H, ArH), 9.84 (s, 1H, NH). <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 118.26, 120.37 (C=C), 121.82, 122.85, 123.41, 124.84, 125.62, 126.84, 127.42, 128.32, 131.64, 133.21, 134.35, 135.68 (Ar C), 179.25 (C=O). Calcd for C<sub>15</sub>H<sub>9</sub>N<sub>2</sub>O<sub>2</sub>: C, 76.60; H, 3.83; N, 5.96%. Found: C, 76.49; H, 3.74, N, 5.87%.

**3'-Substituted-(1',2',4')triazolo[5,6-b][indeno (2,3-e)]-1,3,4-thiadiazine, 7a–c.** An equimolar mixture of compound **2** (0.01 mol) and substituted triazole (0.0 mol) in ethanol (20 mL) was refluxed in presence of DMF (0.02 mol) for about 4–5 h. The progress of reaction was monitored on TLC. Upon completion of reaction, the reaction mixture was quenched into crushed ice. The product precipitated out was filtered, washed with water, and purified by recrystallization from ethanol to yield **7a–c**.

**3'-H-(1',2',4') triazolo [5,6-b][indeno (2,3-e)]-1,3,4-thiadiazine (7a).** M.p. = 215–217°C, Yield = 81%, IR (cm<sup>-1</sup>): 3353, 1730, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 7.09–8.54 (m, 5H, ArH, CH), 9.84 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 122.15, 124.36 (C=C), 126.84, 127.45, 128.14, 130.53, 131.14, 132.92 (Ar C), 148.54 (C=N), 156.56 (C=N), 168.48 (C=O). Calcd for

C<sub>11</sub>H<sub>6</sub>N<sub>4</sub>O<sub>2</sub>: C, 54.55; H, 2.48; N, 23.14%. Found: C, 54.46; H, 2.38; N, 23.01%.

**3'-Methyl-(1',2',4')triazolo [5,6-b][indeno (2,3-e)]-1,3,4-thiadiazine (7b).** M.p. = 201–203°C, Yield = 87%, IR (cm<sup>-1</sup>): 3153, 1730, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 2.42 (s, 3H, CH<sub>3</sub>), 7.15–8.35 (m, 4H, ArH), 9.74 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 23.67 (CH<sub>3</sub>), 123.54, 125.38 (C=C), 128.79, 129.51, 130.12, 131.08, 131.97, 132.41 (Ar C), 149.54 (C=N), 155.98 (C=N), 167.47 (C=O). Calcd for C<sub>12</sub>H<sub>8</sub>N<sub>4</sub>O<sub>2</sub>: C, 56.24; H, 3.15; N, 21.86%. Found: C, 56.34; H, 3.12; N, 21.79%.

**3'-Ethyl-(1',2',4')triazolo[5,6-b][indeno (2,3-e)]-1,3,4-thiadiazine (7c).** M.p. = 201–203°C, Yield = 87%, IR (cm<sup>-1</sup>): 3153, 1730, <sup>1</sup>H NMR (500 MHz, DMSO-*d*<sub>6</sub>, δ ppm): 2.42 (q, 3H, CH<sub>3</sub>), 2.21 (t, 2H, CH<sub>2</sub>), 6.85–7.98 (m, 4H, ArH), 9.68 (s, 1H, NH), <sup>13</sup>C NMR (500 MHz, DMSO-*d*<sub>6</sub>, ppm): 23.67 (CH<sub>3</sub>), 29.48 (CH<sub>2</sub>), 124.53, 127.34 (C=C), 127.82, 127.98, 128.73, 129.01, 130.56, 131.98 (Ar C), 150.55 (C=N), 154.85 (C=N), 170.42 (C=O). Calcd for C<sub>13</sub>H<sub>10</sub>N<sub>4</sub>O<sub>2</sub>: C, 57.78; H, 3.70; N, 20.74%. Found: C, 57.71; H, 3.62; N, 20.68%.

## ANTIMICROBIAL EVALUATION

The newly synthesized compounds were tested for their antimicrobial activity against the following microorganisms: (a) Gram-negative: *Escherichia coli*, *Pseudomonas putide*; (b) Gram-positive: *Bacillus subtilis*, *Streptococcus lactis*; (c) Fungi: *Aspergillus niger*, *Penicillium sp.*; (d) Yeast: *Candida albicans*. The preliminary screening of the investigated compounds was performed using the filter paper disk-diffusion method [14]. The compounds were tested at a concentration of 100 µg/mL. The zone of inhibition was measured in millimeter and compared with reference standard ampicillin trihydrate (100 µg/mL). The compounds tested displayed good activity toward Gram positive bacteria, but were less active against Gram-negative bacteria. The results of antibacterial screening studies are reported in Table 2.

**Table 2**

Antimicrobial activities of some newly synthesized compounds.

Compound	Inhibition zone (mm)						
	Gram negative		Gram positive		Fungi		Yeast
	<i>E. coli</i>	<i>P. putide</i>	<i>B. subtilis</i>	<i>S. lactis</i>	<i>A. niger</i>	<i>P. sp.</i>	<i>C. albicans</i>
<b>3d</b>	17	15	18	21	12	10	5
<b>4f</b>	16	16	17	21	10	10	5
<b>5a</b>	15	14	18	19	8	8	5
<b>5c</b>	18	19	19	20	8	8	5
<b>7b</b>	13	18	17	20	0	0	0
Ampicilin®	24	20	19	22	24	14	14

*E. coli*, *Escherichia coli*; *P. putide*, *Pseudomonas putide*; *B. subtilis*, *Bacillus subtilis*; *S. lactis*, *Streptococcus lactis*; *A. niger*, *Aspergillus niger*; *P. sp.*, *Penicillium sp.*; *C. albicans*, *Candida Albicans*.

The sensitivity of microorganisms to the tested compounds is identified in the following manner:

Highly sensitive = inhibition zone: 15–20 mm.

Moderately sensitive = inhibition zone: 10–15 mm.

Slightly sensitive = inhibition zone: 5–10 mm.

Not sensitive = inhibition zone: 0 mm.

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