



**SYNTHESIS, CHARACTERIZATION, ANTIMICROBIAL,  
ANTIOXIDANT AND LARVICIDAL ACTIVITIES OF NOVEL  
[1-(5,7-DICHLORO-1,3-BENZOXAZOL-2-YL)-3-  
SUBSTITUTED PHENYL-1H-PYRAZOL-4-L]  
[METHYLENE] ANILINE DERIVATIVES**

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**ABSTRACT**

The Schiff base of [1-(5,7-dichloro-1,3-benzoxazol-2-yl)-3-substituted phenyl-1H-pyrazol-4-yl] methylene] aniline (**2a-j**) have been synthesized by condensation reaction of 1-(5,7-dichloro-1,3-benzoxazol-2-yl)-3- substituted phenyl-1H-pyrazole-4-carbaldehyde derivatives with aromatic amines in methanol. The reaction carried out at room temperature on stirring. The target compounds (**2a-j**) were characterized by IR, <sup>1</sup>H NMR, Mass and <sup>13</sup>C NMR spectral analysis, also subjected to antimicrobial, antioxidant and larvicidal activity. The compounds (**2b**), (**2e**) and (**2f**) showed marked antimicrobial activity and compounds (**2b**), (**2a**) and (**2e**) acts as DPPH scavenging agents. The compound (**2c**), (**2d**) and (**2f**) acts as reducing agents. The compounds (**2b**) and (**2c**) showed potent larvicidal activity.

**Key words:** Dichlorobenzoxazole, Anti-oxidant, Larvicidal, Schiff base.

**INTRODUCTION**

The benzoxazoles have been attracted the more researchers for many years because they constitute an important class of heterocyclic compounds<sup>1</sup>. A sequence of analogue and derivatives of heterocyclic compounds containing nitrogen, oxygen and oxazole moieties

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constitutes the core structure of a numerous biologically active compounds. Oxazole containing heterocyclic compounds play an imperative role in medicinal chemistry and exhibit wide range of biological activities<sup>2-5</sup>. There are abundant methods in synthesizing benzoxazole derivatives because the five-membered heterocyclic derivatives have been targeted more than half a century<sup>6,7</sup>. Schiff bases important class of compounds in medicinal and pharmaceutical field. Literature survey shows that many Schiff bases exhibit biological properties include antibacterial, antifungal<sup>8-13</sup> and antitumor activity<sup>14,15</sup>. In recent days, Schiff bases are more frequently applied for the betterment of human welfare. The importance of the Schiff base is due its versatile nature. These observations motivated us to synthesis of Schiff bases at room temperature in the present work. The conception that Schiff bases, [1-(5,7-dichloro-1,3-benzoxazol-2-yl)-3-substituted phenyl-1*H*-pyrazol-4-yl] methylene] aniline (**2a-j**) posses potential antimicrobial, antioxidant and larvicidal activities.

## EXPERIMENTAL

### Chemistry

Melting points were recorded on electro thermal melting point apparatus and are uncorrected. <sup>1</sup>H NMR and <sup>13</sup>C NMR spectra were recorded on Bruker 400 MHz spectrometer in IISc, Bangalore, Karnataka, India. The chemical shifts are shown in  $\delta$  values (ppm) with tetramethylsilane (TMS) as an internal standard. LC-MS were obtained using C 18 column on Shimadzu, LCMS 2010A, Japan. The FT-IR spectra of the compounds were taken in KBr pellet (100 mg) using Shimadzu Fourier Transformed Infrared (FT-IR) Spectrophotometer. The column chromatography was performed using silica gel (230-400 mesh). Silica gel GF254 plates from Merck were used for TLC and spots located either by UV or dipping in potassium permanganate solution. The chemicals were purchased from Sigma-Aldrich Co. and from SD Fine chemicals. The solvents for column chromatography were of reagent grade and were purchased from commercial source.

The preparation of 1-(5, 7-dichloro-1,3-benzoxazol-2-yl)-3-phenyl-1*H*-pyrazole-4-carbaldehyde derivatives **1(a-j)** were reported in our previous work.<sup>16</sup>

### General procedure for preparation of [1-(5,7-dichloro-1,3-benzoxazol-2-yl)-3-substituted phenyl-1*H*-pyrazol-4-yl]methylene}aniline derivatives (**2a-j**)

The compounds (**1a-j**) (0.001 M) and aromatic amines (0.001 M) were taken in methanol and stirred for 4 hr in room temperature. The completion of reaction was checked by TLC. The reaction mass was filtered to get solid product. The solid was dried and recrystallised using ethanol.

2-Chloro-N-{(1E)-[1-(5,7-dichloro-1,3-benzoxazol-2-yl)-3-(4-methoxyphenyl)-1H-pyrazol-4-yl]methylene}aniline (**2a**). Yield-91%, M.P. 185°C, IR (KBr)  $\text{cm}^{-1}$ : 2929 (OCH<sub>3</sub>), 1472 (C=C), 760 (C-Cl); <sup>1</sup>H NMR (DMSO-d<sub>6</sub>) ppm:  $\delta$  3.82 (s, 3H, OCH<sub>3</sub>),  $\delta$  7.07-8.04 (10H, Ar H),  $\delta$  8.6 (s, 1H, pyrazole),  $\delta$  9.18 (s, 1H, -CH=N), <sup>13</sup>C NMR (DMSO-d<sub>6</sub>):, 163.15, 159.19, 156.05, 150.33, 138.49, 136.24, 133.92, 131.45, 128.47, 128.26, 128.04, 127.92, 127.86, 127.74, 126.99, 125.56, 122.93, 121.20, 119.93, 111.24, 110.61, 109.52, 109.01, (23 Ar-C), 55.80 (methoxy), M<sup>+</sup> 499.3, Anal. Calcd for (%) C<sub>24</sub>H<sub>15</sub>Cl<sub>3</sub>N<sub>4</sub>O<sub>2</sub> (497.7 g/mole): C (57.91%) H (3.04%) N (11.26%) Found: C (57.92%) H (3.03%) N (11.25%).

4-[4-{(E)-[(2-Chlorophenyl)imino]methyl}-1-(5,7-dichloro-1,3-benzoxazol-2-yl)-1H-pyrazol-3-yl]phenol (**2b**). Yield-90%, M.P. 178°C, IR (KBr)  $\text{cm}^{-1}$ : 3494 (OH), 1479 (C=C), 769 (C-Cl); <sup>1</sup>H NMR (DMSO-d<sub>6</sub>) ppm:  $\delta$  7.2-8.08 (10 H, Ar H),  $\delta$  8.3 (s, 1H, OH),  $\delta$  8.66 (s, 1H, pyrazole),  $\delta$  9.27 (s, 1H, -CH=N), <sup>13</sup>C NMR (DMSO-d<sub>6</sub>):, 163.20, 159.21, 156.23, 150.15, 138.36, 136.45, 133.81, 131.24, 128.52, 128.56, 128.32, 127.91, 127.82, 127.73, 126.94, 125.55, 122.96, 121.27, 119.94, 111.28, 110.59, 109.56, 109.21, (23 Ar-C), M<sup>+</sup> 485.4, Anal. Calcd for (%) C<sub>23</sub>H<sub>13</sub>Cl<sub>3</sub>N<sub>4</sub>O<sub>2</sub> (483.7 g/mole): C (57.11%) H (2.71%) N (11.58%) Found: C (57.14%) H (2.70%) N (11.56%).

2-Chloro-N-{(1E)-[1-(5,7-dichloro-1,3-benzoxazol-2-yl)-3-(4-methylphenyl)-1H-pyrazol-4-yl]methylene}aniline (**2c**). Yield-87%, M.P. 160°C, IR (KBr)  $\text{cm}^{-1}$ : 3139 (CH<sub>3</sub>), 1466 (C=C), 769 (C-Cl); <sup>1</sup>H NMR (DMSO-d<sub>6</sub>) ppm:  $\delta$  2.5 (s, 3H, CH<sub>3</sub>),  $\delta$  7.1-8.31 (10H, Ar H),  $\delta$  8.64 (s, 1H, pyrazole),  $\delta$  9.22 (s, 1H, -CH=N), <sup>13</sup>C NMR (DMSO-d<sub>6</sub>):, 163.24, 159.15, 156.25, 150.39, 138.25, 136.45, 133.67, 131.44, 128.39, 128.56, 128.92, 127.43, 127.66, 127.64, 126.63, 125.73, 122.84, 121.12, 119.83, 111.20, 110.54, 109.56, 109.44, (23Ar-C), 52.10 (methyl), M<sup>+</sup> 481.7, Anal. Calcd for (%) C<sub>24</sub>H<sub>15</sub>Cl<sub>3</sub>N<sub>4</sub>O (483.2 gm/mole): C (59.83%) H (3.14%) N (11.63%) Found: C (59.81%) H (3.16%) N (11.65%).

N-{(1E)-[3-(4-Bromophenyl)-1-(5,7-dichloro-1,3-benzoxazol-2-yl)-1H-pyrazol-4-yl]methylene}-2-chloroaniline (**2d**). Yield-92%, M.P. 175°C IR (KBr)  $\text{cm}^{-1}$ : 1471 (C=C), 762 (C-Cl); <sup>1</sup>H NMR (DMSO-d<sub>6</sub>) ppm:  $\delta$  7.18-8.24 (10H, Ar H),  $\delta$  8.62 (s, 1H, pyrazole),  $\delta$  9.1 (s, 1H, -CH=N), <sup>13</sup>C NMR (DMSO-d<sub>6</sub>):, 163.23, 159.74, 156.27, 150.45, 138.0, 136.29, 133.82, 131.33, 128.75, 128.56, 128.61, 127, 96, 127.50, 127.64, 126.55, 125.58, 122.71, 121.61, 119.85, 111.58, 110.84, 109.55, 109.74, (23Ar-C), M<sup>+</sup> 548.3. Anal. Calcd for (%) C<sub>23</sub>H<sub>12</sub>BrCl<sub>3</sub>N<sub>4</sub>O (546.6 gm/mole): C (50.54%) H (2.21%) N (10.25%) Found: C (50.52%) H (2.23%) N (10.26%).

2-Chloro-N-{(1E)-[1-(5,7-dichloro-1,3-benzoxazol-2-yl)-3-(3,4-dichlorophenyl)-1H-pyrazol-4-yl]methylene}aniline (**2e**). Yield-84%, M.P. 195°C, IR (KBr)  $\text{cm}^{-1}$ : 1459 (C=C),

768 (C-Cl);  $^1\text{H}$  NMR (DMSO- $d_6$ ) ppm:  $\delta$  7.21-8.29 (9H, Ar H),  $\delta$  8.61 (s, 1H, pyrazole),  $\delta$  9.17 (s, 1H, -CH=N),  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):, 163.25, 159.76, 156.21, 150.35, 138.43, 136.25, 133.62, 131.73, 128.85, 128.46, 128.21, 127.26, 127.20, 127.74, 126.95, 125.88, 122.73, 121.66, 119.65, 111.56, 110.86, 109.56, 109.76, (23Ar-C),  $M^{+2}$  538.3,  $M^{+4}$  540.3, Anal. Calcd for (%)  $\text{C}_{23}\text{H}_{11}\text{Cl}_5\text{N}_4\text{O}$  (536.6 g/mole): C (51.48%) H (2.07%) N (10.44%) Found: C (51.46%) H (2.05%) N (10.42%).

3-Chloro-N-{(1E)-[1-(5, 7-dichloro-1, 3-benzoxazol-2-yl)-3-(4-methoxyphenyl)-1H-pyrazol-4-yl]methylene}aniline (**2f**). Yield-85%, M.P. 169°C, IR (KBr)  $\text{cm}^{-1}$ : 2925 ( $\text{OCH}_3$ ), 1471 (C=C), 765 (C-Cl);  $^1\text{H}$  NMR (DMSO- $d_6$ ) ppm:  $\delta$  3.84 (s, 3H,  $\text{OCH}_3$ ),  $\delta$  7.05-8.06 (10H, Ar H),  $\delta$  8.5 (s, 1H, pyrazole),  $\delta$  9.2 (s, 1H, -CH=N),  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):, 163.14, 159.16, 156.15, 150.32, 138.47, 136.25, 133.93, 131.55, 128.46, 128.36, 128.24, 127.95, 127.84, 127.54, 126.92, 125.55, 122.63, 121.10, 119.94, 111.44, 110.63, 109.42, 109.21, (23Ar-C), 55.81 (methoxy),  $M^{+2}$  499.3, Anal. Calcd for (%)  $\text{C}_{24}\text{H}_{15}\text{Cl}_3\text{N}_4\text{O}_2$  (497.7 gm/mole): C (57.91%) H (3.04%) N (11.26%) Found: C (57.93%) H (3.01%) N (11.25%).

N-{(1E)-[1-(5, 7-Dichloro-1, 3-benzoxazol-2-yl)-3-(4-methoxyphenyl)-1H-pyrazol-4-yl]methylene}-3-nitroaniline (**2g**). Yield-92%, M.P. 144°C IR (KBr)  $\text{cm}^{-1}$ : 2929 ( $\text{OCH}_3$ ), 1465 (C=C), 760 (C-Cl);  $^1\text{H}$  NMR (DMSO- $d_6$ ) ppm:  $\delta$  3.83 (s, 3H,  $\text{OCH}_3$ ),  $\delta$  7.2-8.31 (10H, Ar H),  $\delta$  8.4 (s, 1H, pyrazole),  $\delta$  9.23 (s, 1H, -CH=N),  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):, 162.14, 159.12, 156.14, 150.38, 138.49, 136.21, 133.92, 131.55, 128.45, 128.32, 128.34, 127.65, 127.84, 127.34, 126.52, 125.53, 122.43, 121.15, 119.34, 111.42, 110.62, 109.41, 109.25, (23Ar-C), 55.79 (methoxy),  $M^{+2}$  510.3, Anal. Calcd for (%)  $\text{C}_{24}\text{H}_{15}\text{Cl}_2\text{N}_5\text{O}_4$  (508.3 gm/mole): C (56.71%) H (2.97%) N (13.78%) Found: C (56.70%) H (2.99%) N (13.77%).

N-{(1E)-[1-(5, 7-Dichloro-1, 3-benzoxazol-2-yl)-3-(4-methoxyphenyl)-1H-pyrazol-4-yl]methylene}-3-methylaniline (**2h**). Yield-86%, M.P. 173°C, IR (KBr)  $\text{cm}^{-1}$ : 3139 ( $\text{CH}_3$ ), 3011 ( $\text{OCH}_3$ ), 1466 (C=C), 759 (C-Cl);  $^1\text{H}$  NMR (DMSO- $d_6$ ) ppm:  $\delta$  2.51 (s, 3H,  $\text{CH}_3$ ),  $\delta$  3.91 (s, 3H,  $\text{OCH}_3$ ),  $\delta$  7.16-8.23 (10H, Ar H),  $\delta$  8.45 (s, 1H, pyrazole),  $\delta$  9.24 (s, 1H, -CH=N),  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):, 163.11, 159.12, 156.13, 150.34, 138.45, 136.25, 133.96, 131.57, 128.47, 128.38, 128.28, 127.94, 127.89, 127.55, 126.96, 125.56, 122.65, 121.12, 119.64, 111.46, 110.61, 109.32, 109.11, (23Ar-C), 55.81 (methoxy), 52.0 (methyl),  $M^{+2}$  479.3. Anal. Calcd for (%)  $\text{C}_{25}\text{H}_{18}\text{Cl}_2\text{N}_4\text{O}_2$  (477.3 gm/mole): C (62.90%) H (3.80%) N (11.74%) Found: C (62.92%) H (3.82%) N (11.71%).

N-{(1E)-[1-(5,7-Dichloro-1,3-benzoxazol-2-yl)-3-(4-methoxyphenyl)-1H-pyrazol-4-yl]methylene}-4-methylaniline (**2i**). Yield-87%, M.P. 183°C, IR (KBr)  $\text{cm}^{-1}$ : 3137 ( $\text{CH}_3$ ), 3009 ( $\text{OCH}_3$ ), 1468 (C=C), 760 (C-Cl);  $^1\text{H}$  NMR (DMSO- $d_6$ ) ppm:  $\delta$  2.50 (s, 3H,  $\text{CH}_3$ ),  $\delta$

3.92 (s, 3H, OCH<sub>3</sub>),  $\delta$  7.11-8.21 (10H, Ar H),  $\delta$  8.3 (s, 1H, pyrazole),  $\delta$  9.21 (s, 1H, -CH=N), <sup>13</sup>C NMR (DMSO-d<sub>6</sub>):, 163.10, 159.13, 156.12, 150.33, 138.46, 136.27, 133.94, 131, 55, 128.49, 128.31, 128.23, 127.93, 127.86, 127.55, 126.94, 125.58, 122.67, 121.18, 119.64, 111.45, 110.63, 109.31, 109.12, (23Ar-C), 55.83 (methoxy), 52.11 (methyl), M<sup>+2</sup> 479.3, Anal. Calcd for (%) C<sub>25</sub>H<sub>18</sub>Cl<sub>2</sub>N<sub>4</sub>O<sub>2</sub> (477.3 g/mole): C (62.90%) H (3.80%) N (11.74%) Found: C (62.92%) H (3.82%) N (11.70%).

N-{(1E)-[1-(5,7-Dichloro-1,3-benzoxazol-2-yl)-3-(4-methoxyphenyl)-1H-pyrazol-4-yl]methylene}-4-methoxyaniline (**2j**). Yield-94%, M.P. 197°C, IR (KBr) cm<sup>-1</sup>: 3015 (OCH<sub>3</sub>), 1467 (C=C), 771 (C-Cl); <sup>1</sup>H NMR (DMSO-d<sub>6</sub>) ppm:  $\delta$  3.97 (s, 3H, OCH<sub>3</sub>),  $\delta$  3.99 (s, 3H, OCH<sub>3</sub>),  $\delta$  7.14-8.31 (10H, Ar H),  $\delta$  8.32 (s, 1H, pyrazole),  $\delta$  9.25 (s, 1H, -CH=N), <sup>13</sup>C NMR (DMSO-d<sub>6</sub>):, 163.14, 158.92, 156.13, 150.48, 138.44, 136.51, 133.93, 131,54, 128.46, 128.31, 128.36, 127, 65, 127.84, 127.34, 126.42, 125.55, 122.41, 121.12, 119.37, 111.44, 110.66, 109.47, 109.24, (23Ar-C), 55.8 (methoxy), 55.79 (methoxy), M<sup>+2</sup> 495.3, Anal. Calcd for (%) C<sub>25</sub>H<sub>18</sub>Cl<sub>2</sub>N<sub>4</sub>O<sub>3</sub> (493.3 g/mole): C (60.86%) H (3.68%) N (11.36%) Found: C (60.83%) H (3.67%) N (11.38%).

## Biological activities

### Antibacterial activity

The antibacterial efficacy of compounds was tested against two Gram positive bacteria namely *Staphylococcus aureus* and *Bacillus cereus* and Gram negative bacteria namely *Pseudomonas aeruginosa* and *Escherichia coli* by agar well diffusion method<sup>17</sup>. Twenty four old Muller-Hinton broth cultures of test bacteria were swabbed on sterile Muller-Hinton agar plates using sterile cotton swab followed by punching wells of 6 mm with the help of sterile cork borer. The standard drug (Chloramphenicol, 1 mg/mL of sterile distilled water), compounds (**2a-j**) (20 mg/mL in 10% DMSO) and control (10% DMSO) were added to respectively labeled wells. The plates are allowed to stand for 30 minutes and were incubated at 37°C for 24 h in upright position and the zone of inhibition was recorded.

### Antioxidant activity

#### DPPH assay

The radical scavenging ability of synthesized compounds and the ascorbic acid (standard) was tested on the basis of the radical scavenging effect on DPPH free radical. Different concentration of compounds and standard namely 25, 50, 100, 200 and 400  $\mu$ g/mL were prepared in methanol. In clean and labeled test tubes, 2 mL of DPPH solution (0.002% in methanol) was mixed with 2 mL of different concentration of compounds and standard

separately. The tubes were incubated at room temperature in dark for 30 minutes and the optical density was measured at 517 nm using UV-Visible Spectrophotometer. The absorbance of the DPPH control was also noted. The scavenging activity was calculated using the formula:

Scavenging activity (%) =  $(A - B) / A \times 100$ , where A is the absorbance of DPPH and B is the absorbance of DPPH and in standard combination<sup>18</sup>.

### Ferric reducing assay

Different concentration of compounds and standard (tannic acid) namely 25, 50, 100, 200 and 400  $\mu\text{g/mL}$  in 1 mL of methanol were mixed in separate tubes with 2.5 mL of phosphate buffer (200 mM, pH 6.6) and 2.5 mL of 1% potassium ferricyanide. The tubes were placed in water bath for 20 minutes at 50°C, cooled rapidly and mixed with 2.5 mL of 10% trichloroacetic acid and 0.5 mL of 0.1% ferric chloride. The amount of iron (II)-ferricyanide complex formed was determined by measuring the formation of Perl's Prussian blue at 700 nm after 10 minutes. The increase in absorbance of the reaction mixture indicated the increased reducing power<sup>19</sup>.

### Larvicidal activity

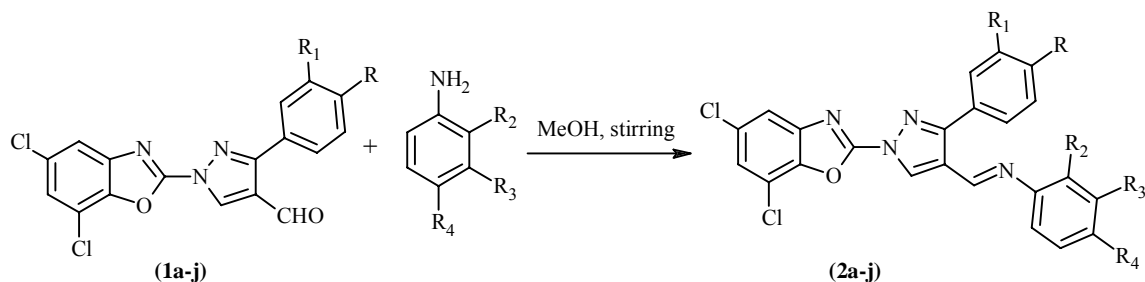
Insecticidal activity of the compounds (**2a-j**) was tested against second and third instar larvae of *Aedes aegypti* mosquito. The concentration of the compounds (0.5 mg/mL) was prepared in 10% DMSO and added to sterile labeled beakers containing 25 mL of water. Twenty larvae were placed in each of the beakers containing compounds (**2a-j**). A control was kept in 10% DMSO. The insecticidal effect was determined by counting the number of dead larvae after 24 hours. Dead larvae were identified when they failed to move after probing with a needle in siphon or cervical region. Each test was repeated thrice; the percentage of larval mortality was determined<sup>20</sup>.

## RESULTS AND DISCUSSION

### Chemistry

In the present study, the synthesis of Schiff bases is reported by the reaction with 3-(4-methoxyphenyl)-1-substituted phenyl-1*H*-pyrazole-4-carbaldehyde derivatives and 3-chloro-4-fluoroaniline via condensation reaction at room temperature in excellent yield (**Scheme 1**). In most of the cases Schiff bases were synthesized upon heating or on reflux. The functional group transformation of compounds (**1a-j**) into compounds (**2a-j**) was established on the basis of IR, <sup>1</sup>H NMR and <sup>13</sup>C NMR and mass spectral data.

In the  $^1\text{H}$  NMR spectrum of compound (**1a-j**), the aldehydic proton signal at 9.95-9.997 ppm disappeared and a new signal of =C-NH- arose in the region of 8.62-8.65 signal. Moreover, in the  $^{13}\text{C}$  NMR spectrum the aldehydic carbon of compound (**1a-j**), after transforming into =C-NH showed a new carbon signal at 159.12-159.51 ppm. The complete spectral details of compounds (**2a-j**) are mentioned in the experimental part. The values are in complete agreement with the structure assigned.



Scheme 1

Compound	R	R <sub>1</sub>	R <sub>2</sub>	R <sub>3</sub>	R <sub>4</sub>
<b>2a</b>	OCH <sub>3</sub>	H	Cl	H	H
<b>2b</b>	OH	H	Cl	H	H
<b>2c</b>	CH <sub>3</sub>	H	Cl	H	H
<b>2d</b>	Br	H	Cl	H	H
<b>2e</b>	Cl	Cl	Cl	H	H
<b>2f</b>	OCH <sub>3</sub>	H	H	Cl	H
<b>2g</b>	OCH <sub>3</sub>	H	H	NO <sub>2</sub>	H
<b>2h</b>	OCH <sub>3</sub>	H	H	CH <sub>3</sub>	H
<b>2i</b>	OCH <sub>3</sub>	H	H	H	CH <sub>3</sub>
<b>2j</b>	OCH <sub>3</sub>	H	H	H	OCH <sub>3</sub>

### Biological evaluation

The result of antibacterial activity of compounds (**2a-j**) is shown in Table 1. In the antibacterial study, the compounds have shown inhibition of test bacteria. Among the compounds, marked inhibition of test bacteria was observed in compounds (**2b**), (**2e**) and (**2f**) as compared with standard drugs.

**Table 1: Antibacterial activity of compounds 2(a-j)**

Compound	Zone of inhibition in (mm), concentration ( $\mu\text{g/mL}$ )							
	<i>S. aureus</i>		<i>B. subtilis</i>		<i>E. coli</i>		<i>P. aeruginosa</i>	
	20	10	20	10	20	10	20	10
<b>2a</b>	12	10	13	10	10	---	12	09
<b>2b</b>	16	13	12	08	08	---	12	08
<b>2c</b>	12	09	11	08	08	---	09	---
<b>2d</b>	11	08	10	---	08	---	09	---
<b>2e</b>	14	10	11	08	08	---	08	---
<b>2f</b>	14	11	10	08	08	---	08	---
<b>2g</b>	11	09	09	08	08	---	07	---
<b>2h</b>	10	08	08	07	08	---	08	---
<b>2i</b>	11	08	10	08	07	---	07	08
<b>2j</b>	10	09	08	09	08	---	08	08
<b>DMSO</b>	00	00	00	00	00	00	00	00
<b>Standard</b>	34	34	35	35	32	32	30	30

DMSO- Dimethyl sulphoxide

Standard- Chloremphenicol

Antioxidant activity of different concentration of compounds (**2a-j**) in methanol and ascorbic acid in terms of free radical scavenging ability was evaluated using DPPH free radical assay. The compounds exhibited marked antioxidant activity by scavenging DPPH\* (free radical) and converting into DPPH and the activity was found to be dose dependent. The compound (**2b**) was showed more potent followed by compounds (**2a**) and (**2e**). The results were tabulated in Table 2.

**Table 2: DPPH free radical scavenging activity of compounds 2(a-j)**

Compds.	Scavenging activity of different concentrations ( $\mu\text{g/mL}$ ) of compounds %				
	400	200	100	50	25
<b>2a</b>	71.65	63.14	51.66	43.14	36.13
<b>2b</b>	73.14	61.69	52.13	40.16	38.14

Cont...



Compds.	Scavenging activity of different concentrations ( $\mu\text{g/mL}$ ) of compounds %				
	400	200	100	50	25
<b>2c</b>	63.14	63.15	53.14	45.14	38.16
<b>2d</b>	65.69	55.14	48.39	42.14	35.16
<b>2e</b>	70.16	64.60	55.39	49.13	37.69
<b>2f</b>	66.46	58.14	49.13	38.36	30.69
<b>2g</b>	62.56	56.11	47.37	37.44	30.21
<b>2h</b>	61.77	56.12	46.20	37.19	30.61
<b>2i</b>	63.01	57.48	47.38	38.87	32.67
<b>2j</b>	64.57	58.09	48.32	37.57	33.00
<b>A A</b>	96.36	93.18	91.56	86.36	81.16

A A-Ascorbic Acid

The result of reducing power of different concentrations of compounds (**2a-j**) and tannic acid is represented in Table 3. In this study, the absorbance was found to be increased with the dose of compounds and standard which is suggestive reducing power. The compounds (**2c**), (**2d**) and (**2f**) were showed potent reducing power.

**Table 3: Ferric reducing activity of compounds 2(a-j)**

Compounds	Absorbance at 700 nm				
	400	200	100	50	25
<b>2a</b>	0.68	0.53	0.41	0.35	0.31
<b>2b</b>	0.51	0.45	0.39	0.30	0.25
<b>2c</b>	0.63	0.55	0.46	0.41	0.39
<b>2d</b>	0.58	0.56	0.50	0.45	0.43
<b>2e</b>	0.55	0.50	0.45	0.40	0.38
<b>2f</b>	0.69	0.65	0.59	0.56	0.54
<b>2g</b>	0.50	0.48	0.40	0.31	0.24
<b>2h</b>	0.52	0.44	0.38	0.33	0.23
<b>2i</b>	0.54	0.41	0.35	0.32	0.26
<b>2j</b>	0.55	0.43	0.34	0.30	0.23
<b>Tannic acid</b>	1.24	1.18	1.11	1.02	0.89

The compounds (**2a-j**) were subjected to insecticidal activity to know the mortality of the compounds against *Aedes aegypti* mosquito larvae. The compound (**2b**) and (**2c**) exhibited higher mortality as compared to other synthesized compounds. The results were tabulated in Table 4.

**Table 4: Larvicidal activity of compounds 2(a-j)**

Compounds	% Mortality of larvae, concentration ( $\mu\text{g/mL}$ )			
	24 hrs		48 hrs	
	10	20	10	20
<b>2a</b>	25	35	50	65
<b>2b</b>	45	75	70	85
<b>2c</b>	30	60	50	75
<b>2d</b>	15	35	35	50
<b>2e</b>	25	40	60	65
<b>2f</b>	15	40	45	60
<b>2g</b>	25	30	35	45
<b>2h</b>	15	35	50	55
<b>2i</b>	25	35	40	50
<b>2j</b>	15	30	35	50
<b>Standard</b>	100	100	100	100
Standard-Melathione				

## CONCLUSION

This study reports the successful synthesis of the title compounds at room temperature in good yield and characterized by IR,  $^1\text{H}$  NMR, Mass and  $^{13}\text{C}$  NMR spectral analysis. The synthesized target molecules screened for some biological activities. The compounds (**2b**), (**2e**), and (**2f**) showed marked antimicrobial activity and compounds (**2b**), (**2a**) and (**2e**) exhibited potent DPPH scavenging activity, whereas compounds (**2c**), (**2d**) and (**2f**) act as reducing agents and that were confirmed by antioxidant activity. Among the synthesized compounds, the compounds (**2b**) and (**2c**) showed higher larvicidal activity. Considering the data and results obtained, it can be concluded that, some derivatives of the

Schiff's bases of [1-(5,7-dichloro-1, 3-benzoxazol-2-yl)-3-phenyl-1*H*-pyrazol-4-yl]methylene] aniline-derivatives were found to be biologically potent heterocycles.

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### REFERENCES

1. E. Tugba, Y. Ilkay, T-G. Betul, B. Kayhan, T.-A. Ozlem, O. Semiha, K. Fatma, Y. Ismail and A. Esin, *Eur. J. Med. Chem.*, **1** (2008).
2. C. Ramalingan, S. Balasubramanian, S. Kabilan and M. Vasudevan, *Eur. J. Med. Chem.*, **39**, 527 (2004).
3. G. Turan-Zitouni, S. Demirayak, A. Ozdemir, Z. A. Kaplacikli and M. T. Yildiz, *Eur. Med. Chem.*, **39**, 267 (2003).
4. O. Temiz, I. Oren, E. A. Sener, I. Yalcin and N. Ucartuerk, *Farmaco*, **53**, 337 (1998).
5. I. Oren, O. Temiz, I. Yalcin, E. A. Sener and N. Altanlar, *Eur. J. Pharm. Sci.*, **7**, 153 (1998).
6. J. B. Wright, *Chem. Rev.*, **48**, 397 (1951).
7. P. N. Preston, *Chem. Rev.*, **74**, 279 (1974).
8. A. A. A. Abu-Hussen, *J. Coord. Chem.*, **59**, 157 (2006).
9. M. Sithambaram Karthikeyan, D. Jagadesh Prasad, B. Poojary and K. Subramanya Bhat, *Bioorg. Med. Chem.*, **14**, 7482 (2006).
10. K. Singh, M. S. Barwa and P. Tyagi, *Eur. J. Med. Chem.*, **41**, 147 (2006).
11. R. R. Pannerselvam, R. R. Nair, G. Vijayalakshmi, E. H. Subramanian and S. K. Sridhar, *Eur. J. Med. Chem.*, **40**, 225 (2005).
12. S. K. Sridhar, M. Saravan and A. Ramesh A, *Eur. J. Med. Chem.*, **36**, 615 (2001).
13. S. N. Pandeya, D. Sriram, G. Nath and E. Declercq, *Eur. J. Pharmacol.*, **9**, 25 (1999).
14. R. Mladenova, M. Ignatova, N. Manolova, T. Petrova and I. Rashkov, *Eur. Polym. J.*, **38**, 989 (2002).

15. O. M. Walsh, M. J. Meegan, R. M. Prendergast and T. A. Nakib, *Eur. J. Med. Chem.*, **31**, 989 (1996).
16. N. D. Jayanna, H. M. Vagdevi, J. C. Dharshan and T. R. Prashith Kekuda, International Conference on Synthetic & Structural Chemistry, Dec. 8-10 (2011).
17. B. Tepe, E. Donmez, M. Unlu, F. Candan, D. Daferera, G. Vardar-Unlu, M. Polissiou and A. Sokmen, *Food Chemistry*, **84(4)**, 519 (2004).
18. Y. S. Ravikumar, K. M. Mahadevan, M. N. Kumaraswamy, V. P. Vaidya, H. Manjunatha, V. Kumar and N. D. Satyanarayana, *Environmental Toxicology and Pharmacology*, **26**, 142 (2008).
19. M. Oyaizu, *Jpn. J. Nutr.*, **44**, 307 (1986).
20. T. R. Prashith Kekuda, K. S. Vinayaka, N. Mallikarjun, A. C. Bharath, B. Shailendra Kumar, M. C. Rakesh Kumar and H. R. Vinod Kumar, *Pharmacognosy J.*, **2(18)**, 65 (2011).

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