



## Organyltellurium(IV) Bidentate Schiff Base Metal Complexes: Synthesis, Characterization and Biological Activities

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Condensation of 5-(hydroxymethyl)furan-2-carboxaldehyde with 4-toluidine yielded a Schiff base (5-((p-tolylimino)methyl)furan-2-yl). A series of seven novel hexa-coordinated organytellurium(IV) complexes of type  $\text{TeCl}_4 \cdot \text{HMeFPT}$ ,  $\text{RTeCl}_3 \cdot \text{HMeFPT}$  and  $\text{R}_2\text{TeCl}_2 \cdot \text{HMeFPT}$  (where R belongs to 4-methoxyphenyl, 4-hydroxyphenyl and 3-methyl-4-hydroxyphenyl; L belongs to HMeFPT *i.e.* Schiff base have been prepared and characterized by molar conductance, IR, UV,  $^1\text{H}$  NMR,  $^{13}\text{C}$  NMR, mass spectroscopy and elemental analysis. Thus according to spectroscopic studies, Schiff base ligand functions as a NO donor bidentate ligand across all organytellurium(IV) complexes *via* the azomethine nitrogen atom and the oxygen atom of the furan ring. The above mentioned spectroscopic analysis indicated that all the organytellurium(IV) complexes have distorted octahedral geometry. Schiff base and all the organytellurium(IV) complexes had their geometry optimized and theoretical quantum mechanical characteristics computed. The octahedral geometry for complexes is also suggested by this computing analysis. The antimicrobial property of the Schiff base ligand and its organytellurium(IV) complexes was evaluated *in vitro* against different bacterial strains *viz.* *A. clavatus*, *C. albicans*, *A. niger*, *S. pyogenes*, *S. aureus*, *E. coli* and *P. aeruginosa*. Antimicrobial property of heterocyclic bidentate Schiff base and its organytellurium(IV) complexes was found to be significantly greater than that of some standard antibiotics.

**Keywords:** 5-(Hydroxymethyl)furan-2-carboxaldehyde, 4-Toluidine, Organyltellurium, Antimicrobial activity.

### INTRODUCTION

The chemistry of Schiff base ligands and its corresponding metal complexes has vastly increased, encompassing a wide range of organometallic compounds and bioinorganic chemistry [1]. They are regarded as privileged ligands because Schiff base ligands are primarily produced through condensation between aldehydes and primary amines [2] and are also classified [3] as azomethines, imines or anils. The azomethine group ( $\text{R}_1\text{R}_2\text{C}=\text{NR}_3$ ) is a prominent structural group of Schiff bases, with substituents that might be an alkyl, cycloalkyl, aryl or heterocyclic group [4,5]. Schiff bases are a class of organic compounds that is regarded to be particularly significant and have been investigated extensively in past few years and have attracted a lot of attention due to their appealing chemical and physical features. They are versatile and have a broad range of applications, such as oxidation of olefins [6-10] and catalysts in hydrogenation, photochromic industries [11] and fluorescence sensors for

hazardous metal ions [12]. The advancement of coordination chemistry [13-15] was further aided by Schiff base compounds. Their antiviral [16], antimicrobial [17], antitumors [18], anticancer [19,20] activities of Schiff bases have been reported.

Antibacterial materials are becoming more widely used in a variety of applications, including medical equipment, health care, hygiene applications, water purification systems, health center, dental surgery equipment, textiles, packaging materials and storage [21,22] as public healthcare awareness of the pathogenic impacts, malodors and stain formations induced by microorganisms growth. As a result, the biological activity of Schiff bases requires additional research. As furan [23-25] ring containing heterocyclic atom exhibits to possess distinguish biological activities like antitumor, anti-inflammatory, antimicrobial, antiviral, *etc.*

Tellurium tetrachloride and organytellurium(IV) chlorides ( $\text{RTeCl}_3/\text{R}_2\text{TeCl}_2$ ) have been shown to act as Lewis acids, accepting electron pairs from various sulphur, oxygen and nitrogen donor

atoms in the different Schiff base ligand and forming robust coordination compounds. We focused our research on the synthesis, structural characterization and biological activities of Schiff base (5-((*p*-tolylimino)methyl)furan-2-ol) and complexes, as well as computational investigations.

## EXPERIMENTAL

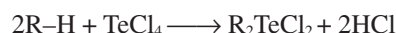
All of the chemicals involved were of the finest standard. Sigma-Aldrich provided tellurium tetrachloride, 4-toluidine, 5-(hydroxymethyl)furan-2-carboxaldehyde, 4-methoxyphenyl, 4-hydroxyphenyl and 3-methyl-4-hydroxyphenyl. The organic solvents were purified using standard procedures. The solvents were kept on molecular sieves for storage. The compounds organytellurium(IV) trichlorides and diorganyltellurium (IV) dichlorides were prepared in a dry N<sub>2</sub> environment.

All melting points values of (HMeFPT) ligand as well as its complexes were calculated *via* capillaries and thus are uncorrected. On FT-infrared spectrophotometer (NICOLET iS50), FT-infrared spectra of HMeFPT and organytellurium(IV) complexes were obtained in the mid IR spectral region in KBr pellets and in the far IR spectral region in polyethylene. The mass spectra of Schiff base (HMeFPT) ligand and its organytellurium(IV) complexes were determined on SCIEX Triple TOF5600 mass spectrophotometer. A conductivity cell containing 10<sup>-3</sup> M solutions in DMSO was used to quantify the molar conductance of all complexes at 25 ± 1 °C. Thermo-Finnigan CHNS analyzer was used for the CHN analyses. The UV-Vis spectra were obtained using a Shimadzu UV-3600 Plus and TMS was used as a reference in DMSO to record <sup>1</sup>H and <sup>13</sup>C NMR with a 400 MHz NMR Spectrometer (BRUKER advance III).

**Synthesis of Schiff base(5-((*p*-tolylimino)methyl)furan-2-ol) (HMeFPT) ligand:** 5-(Hydroxymethyl)furan-2-carboxaldehyde (3.77 g) was mixed using 25 mL of ethanol. Then solution was mixed with a 4-toluidine ethanolic solution (3.21 g). Following complete addition, the reaction mixture was stirred for 4 h on a hot water bath with continuous shaking. Extracting a red solid, recrystallized using ethanol and vacuum dried (**Scheme-I**). *m.w.*: 201.22 g/mol; *m.p.*: 141-144 °C; *yield*: 82%; *Colour*: yellow; <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ (ppm): 8.51 (s, 1H, CH=N), 6.24-7.80 (m, 6H, Ar-H of furan and benzene ring), 2.52 (s, 3H, CH<sub>3</sub>), 9.76 (s, 1H, OH of furanyl ring); <sup>13</sup>C NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ (ppm): 139.49 (C<sub>2</sub>), 115.97 (C<sub>3</sub>), 115.42 (C<sub>4</sub>), 124.61 (C<sub>5</sub>), 146.39 (C<sub>6</sub>), 131.44 (C<sub>7</sub>), 133.24 (C<sub>8,12</sub>), 132.15 (C<sub>9,11</sub>), 131.62 (C<sub>10</sub>), 26.54 (C<sub>13</sub>). Elemental analysis for ligand (C<sub>12</sub>H<sub>11</sub>NO<sub>2</sub>) calcd. (found): C, 71.63 (71.76); H, 5.51 (5.79); N, 6.96 (7.10).

**Synthesis of organytellurium(IV) trichlorides (R<sub>3</sub>TeCl<sub>3</sub>) and diorganyltellurium(IV) dichlorides (R<sub>2</sub>TeCl<sub>2</sub>):** As described in the literature [26-28], electrophilic substitution

reactions of TeCl<sub>4</sub> using 4-methoxyphenyl, 4-hydroxyphenyl and 3-methyl-4-hydroxyphenyl in (1:1 and 1:2) molar ratio resulted in the synthesis of RTeCl<sub>3</sub> and R<sub>2</sub>TeCl<sub>2</sub>.

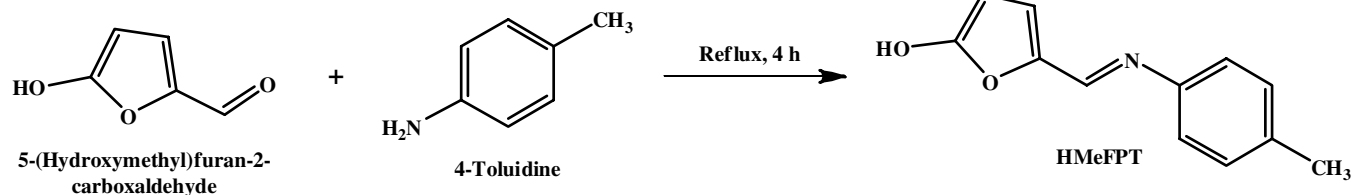


**Synthesis of organytellurium(IV) complexes (TeCl<sub>4</sub>·HMeFPT, RTeCl<sub>3</sub>·HMeFPT and R<sub>2</sub>TeCl<sub>2</sub>·HMeFPT):** The complexes were synthesized by mixing a 20 mL of methanolic solution of ligand HMeFPT (0.01 mol) with 20 mL methanolic solutions of (0.01 mol) TeCl<sub>4</sub>/RTeCl<sub>3</sub>/R<sub>2</sub>TeCl<sub>2</sub> and refluxing at roughly 80 °C for 4 h on a magnetic stirrer. The surplus solvent then distilled and kept for 3-4 days at room temperature. The coloured complexes were then recrystallized in methanol and dried over anhydrous CaCl<sub>2</sub> until their further use (**Scheme-II**).

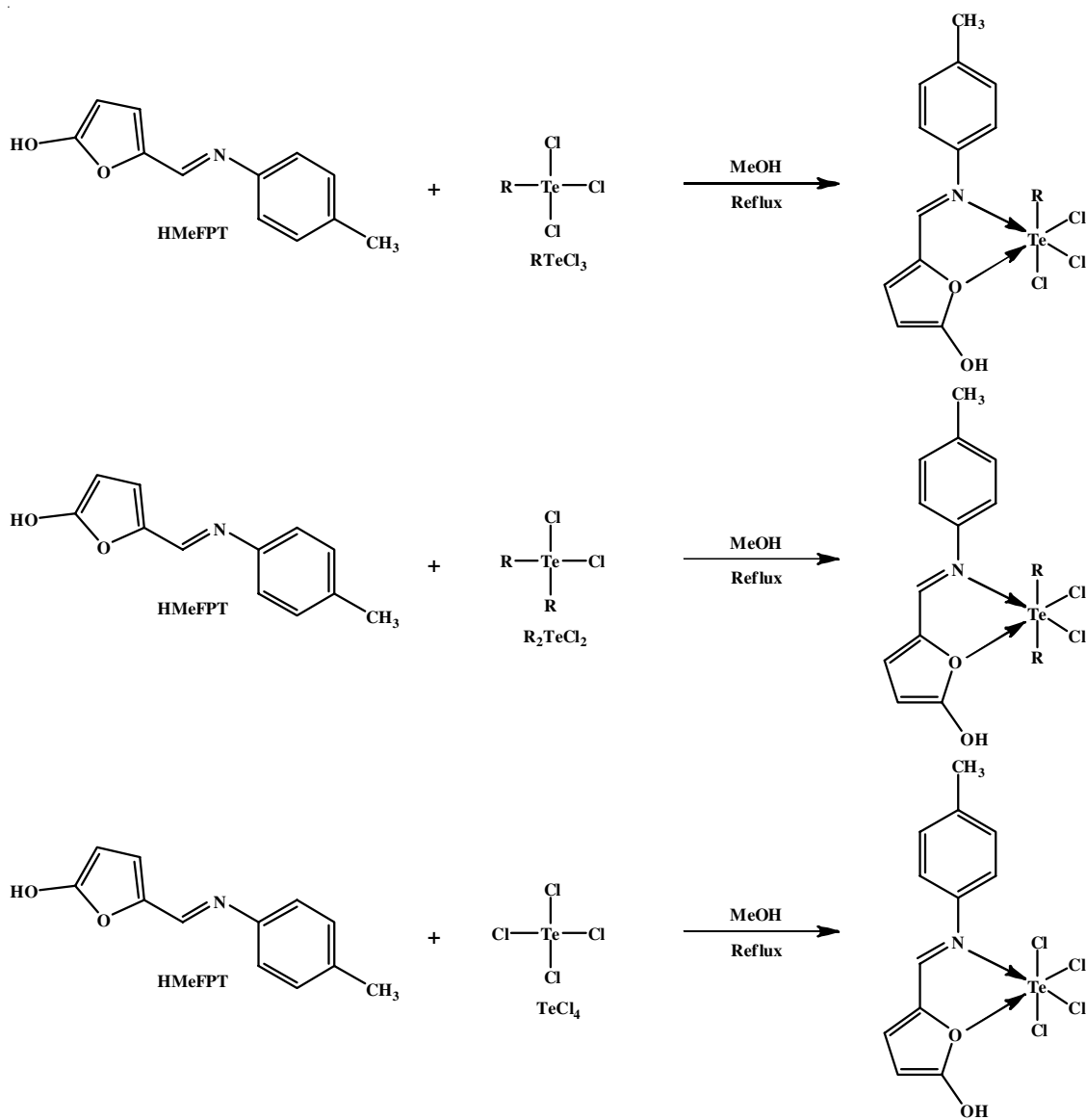
**(4-Hydroxyphenyl) RTeCl<sub>3</sub>·HMeFPT (2a):** *m.w.*: 528.28 g/mol; *m.p.*: 128-136 °C; *yield*: 82.39%; *colour*: yellow; <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ (ppm): 8.67 (s, 1H, C=N), 9.73 (s, 1H, OH of furan ring), 6.27-7.84 (m, 10 H, Ar-H of furan and benzene ring), 10.57 (s, 1H, Ar-OH), 2.53 (s, 3H, CH<sub>3</sub>); <sup>13</sup>C NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ (ppm): 145.36 (C<sub>2</sub>), 109.48 (C<sub>3</sub>), 110.63 (C<sub>4</sub>), 145.28 (C<sub>5</sub>), 165.42 (C<sub>6</sub>), 159.32 (C<sub>7</sub>), 124.36 (C<sub>8,C12</sub>), 136.26 (C<sub>9,C11</sub>), 136.92 (C<sub>10</sub>), 22.51 (C<sub>13</sub>), 141.54 (C<sub>14</sub>), 139.41 (C<sub>15,19</sub>), 137.46 (C<sub>16,18</sub>), 153.28 (C<sub>17</sub>). Elemental analysis for complex **2a** (C<sub>18</sub>H<sub>16</sub>NO<sub>3</sub>Cl<sub>3</sub>Te): calcd. (found) %: C, 40.92 (40.89); H, 3.05 (3.52); N, 2.65 (2.91); Te, 24.15 (24.03); Cl, 20.13 (20.07).

**(4-Methoxyphenyl)RTeCl<sub>3</sub>·HMeFPT (2b):** *m.w.*: 542.94 g/mol; *m.p.*: 127-130 °C; *yield*: 85.92%; *colour*: brown; <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ (ppm): 8.97 (s, 1H, C=N), 9.76 (s, 1H, OH of furan ring), 6.80-7.69 (m, 10 H, Ar-H of furan and benzene ring), 3.94 (s, 3H, OCH<sub>3</sub>), 2.50 (s, 3H, CH<sub>3</sub>); <sup>13</sup>C NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ (ppm): 143.56 (C<sub>2</sub>), 111.52 (C<sub>3</sub>), 109.88 (C<sub>4</sub>), 146.51 (C<sub>5</sub>), 167.23 (C<sub>6</sub>), 161.04 (C<sub>7</sub>), 123.98 (C<sub>8,C12</sub>), 137.79 (C<sub>9,C11</sub>), 139.62 (C<sub>10</sub>), 21.63 (C<sub>13</sub>), 143.29 (C<sub>14</sub>), 140.35 (C<sub>15,19</sub>), 137.25 (C<sub>16,18</sub>), 153.09 (C<sub>17</sub>), 54.79 (C<sub>20</sub>). Elemental analysis for complex **2b** (C<sub>19</sub>H<sub>18</sub>NO<sub>3</sub>Cl<sub>3</sub>Te): calcd. (found) %: C, 42.08 (41.89); H, 3.35 (3.86); N, 2.58 (2.85); Te, 23.53 (22.97); Cl, 19.61 (19.34).

**(3-Methyl-4-hydroxyphenyl)RTeCl<sub>3</sub>·HMeFPT (2c):** *m.w.*: 542.94 g/mol; *m.p.*: 115-119 °C; *yield*: 79.33%; *colour*: brown; <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ (ppm): 8.61 (s, 1H, C=N), 9.77 (s, 1H, OH of furan ring), 6.91-7.88 (m, 10 H, Ar-H of furan and benzene ring), 2.38-2.51 (s, 6H, 2 CH<sub>3</sub>); 9.43 (s, 1H, Ar-OH); <sup>13</sup>C NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ (ppm): 146.35 (C<sub>2</sub>), 111.52 (C<sub>3</sub>), 109.24 (C<sub>4</sub>), 148.35 (C<sub>5</sub>), 167.34 (C<sub>6</sub>), 158.25 (C<sub>7</sub>), 125.63 (C<sub>8,C12</sub>), 140.53 (C<sub>9,C11</sub>), 138.96 (C<sub>10</sub>), 22.14 (C<sub>13</sub>), 144.62 (C<sub>14</sub>), 141.73 (C<sub>15,19</sub>), 135.40 (C<sub>16,18</sub>), 157.89



Scheme-I: Synthesis of Schiff base (HMeFPT) ligand



**Scheme-II:** Synthesis of  $R\text{TeCl}_3\cdot\text{HMeFPT}$ ,  $R_2\text{TeCl}_2\cdot\text{HMeFPT}$  and  $\text{TeCl}_4\cdot\text{HMeFPT}$

(C17), 54.97 (C20). Elemental analysis for complex **2c** ( $\text{C}_{19}\text{H}_{18}\text{NO}_3\text{Cl}_3\text{Te}$ ): calcd. (found)%: C, 42.08 (41.89); H, 3.35 (3.26); N, 2.58 (2.35); Te, 23.53 (23.17); Cl, 19.61 (19.22).

**(4-Hydroxyphenyl) $R_2\text{TeCl}_2\cdot\text{HMeFPT}$  (2d):** *m.w.*: 585.93 g/mol; m.p.: 102-105 °C; yield: 86.15%; colour: brown;  $^1\text{H}$ NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  ppm: 8.56 (s, 1H, C=N), 9.72 (s, 1H, OH of furan ring), 6.76-7.89 (m, 14 H, Ar-H of furan and benzene ring), 2.49 (s, 3H,  $\text{CH}_3$ ); 9.61 (s, 1H, Ar-OH);  $^{13}\text{C}$ NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  ppm: 158.03 (C2), 112.28 (C3), 110.79 (C4), 153.36 (C5), 165.39 (C6), 143.71 (C7), 124.58 (C8,C12), 127.42 (C9,C11), 133.78 (C10), 24.68 (C13), 129.46 (C14), 134.98 (C15,19), 124.14 (C16,18), 154.24 (C17), 127.46 (C20), 135.27 (C21,C25), 124.91 (C22,C24), 153.72 (C23). Elemental analysis for complex **2d** ( $\text{C}_{24}\text{H}_{21}\text{NO}_4\text{Cl}_2\text{Te}$ ): calcd. (found)%: C, 49.20 (49.19); H, 3.61 (3.63); N, 2.39 (2.35); Te, 21.78 (21.74); Cl, 12.10 (12.13).

**(4-Methoxyphenyl) $R_2\text{TeCl}_2\cdot\text{HMeFPT}$  (2e):** *m.w.*: 613.99 g/mol; m.p.: 92-96 °C; yield: 81.49%; colour: dark brown;  $^1\text{H}$ NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  ppm: 8.83 (s, 1H, C=N), 9.76

(s, 1H, OH of furan ring), 6.85-7.91 (m, 14 H, Ar-H of furan and benzene ring), 2.55 (s, 3H,  $\text{CH}_3$ ); 3.87 (s, 3H,  $\text{OCH}_3$ );  $^{13}\text{C}$ NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  ppm: 153.85 (C2), 111.38 (C3), 109.25 (C4), 143.71 (C5), 163.76 (C6), 145.21 (C7), 123.58 (C8,C12), 126.14 (C9,C11), 132.83 (C10), 25.03 (C13), 127.58 (C14), 133.41 (C15,19), 128.35 (C16,18), 140.61 (C17), 124.29 (C20), 55.16 (C21), 134.52 (C22, C26), 129.83 (C23, C25), 140.34 (C24), 54.87 (C27). Elemental analysis for complex **2e** ( $\text{C}_{26}\text{H}_{25}\text{NO}_4\text{Cl}_2\text{Te}$ ): calcd. (found)%: C, 50.86 (50.84); H, 4.10 (4.13); N, 2.28 (2.23); Te, 20.78 (20.75); Cl, 11.55 (11.58).

**(3-Methyl-4-hydroxyphenyl) $R_2\text{TeCl}_2\cdot\text{HMeFPT}$  (2f):** *m.w.*: 613.99 g/mol; m.p.: 137-142 °C; yield: 76.07%; colour: brown;  $^1\text{H}$ NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  ppm: 8.83 (s, 1H, C=N), 9.76 (s, 1H, OH of furan ring), 6.85-7.91 (m, 14 H, Ar-H of furan and benzene ring), 2.55 (s, 3H,  $\text{CH}_3$ ); 3.87 (s, 3H,  $\text{OCH}_3$ );  $^{13}\text{C}$ NMR (400 MHz,  $\text{DMSO-}d_6$ )  $\delta$  (ppm): 147.51 (C2), 108.86 (C3), 109.32 (C4), 142.46 (C5), 167.81 (C6), 143.52 (C7), 124.47 (C8,C12), 127.18 (C9,C11), 131.26 (C10), 25.71 (C13), 128.67 (C14), 132.48 (C15,19), 127.85 (C16,18), 139.76 (C17), 124.72

(C20), 55.24 (C21), 132.62 (C22,C26), 128.32 (C23,C25), 139.26 (C24), 54.58 (C27). Elemental analysis for complex **2f** (C<sub>26</sub>H<sub>25</sub>NO<sub>4</sub>Cl<sub>2</sub>Te): calcd. (found)%: C, 50.86 (50.78); H, 4.10 (4.12); N, 2.28 (2.18); Te, 20.78 (20.69); Cl, 11.55 (11.59).

**(Tellurium tetrachloride)TeCl<sub>4</sub>·HMeFPT (2g):** *m.w.*: 436.19 g/mol; *m.p.*: 141-144 °C; *yield*: 82.28 %; *Colour*: light yellow; <sup>1</sup>H NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ ppm: 8.73 (s, 1H, CH=N), 6.53-7.79 (m, 6H, Ar-H of furan and benzene ring), 2.47 (s, 3H, CH<sub>3</sub>), 9.73 (s, 1H, OH of furanyl ring); <sup>13</sup>C NMR (400 MHz, DMSO-*d*<sub>6</sub>) δ ppm: 146.73 (C2), 106.81 (C3), 106.27 (C4), 142.81 (C5), 165.29 (C6), 134.14 (C7), 132.48 (C8,C12), 131.68 (C9,C11), 132.62 (C10), 25.61 (C13). Elemental analysis for complex **2g** (C<sub>12</sub>H<sub>12</sub>NO<sub>2</sub>Cl<sub>3</sub>Te) calcd. (found)%: C, 33.04 (32.81); H, 2.77 (2.54); N, 3.21 (3.27); Te, 29.25 (29.01); Cl, 24.38 (24.13).

**Computational method:** All DFT computations were represented using the Avogadro and Orca 4.0 software package. The optimization of structures has been done using molecular modelling. The def2-SVP basis sets for the Te(IV) centre and other organic elements (C, H, O and N) were used in the DFT functional to estimate the energies of molecular complexes.

**In vitro antimicrobial activity:** Ligand (HMeFPT) and its organytellurium(IV) complexes (**2a-f**) were screened against fungal strains (*C. albicans* MTCC 227, *A. clavatus* MTCC 1323, *A. niger* MTCC 282), Gram-positive bacterial strains (*S. pyogenes* MTCC 442, *S. aureus* MTCC 96) and Gram-negative bacterial strains (*E. coli* MTCC 443, *P. aeruginosa* MTCC 1688). All of the strains were provided by Institute of Microbial Technology, Chandigarh, India. The antifungal [29] and antibacterial [30,31] activities were evaluated by using broth dilution and the agar well-diffusion methods. DMSO has been used as a diluent to obtain the appropriate drug concentration for testing on standard bacterial strains. At 37 °C, the samples were incubated for 24 to 30 h.

## RESULTS AND DISCUSSION

The refluxing technique was used to synthesize all the tellurium metal Schiff base complexes. The metal complexes were robust, air stable and soluble in ethanol, methanol, DMSO and DMF. The change in colour from ligand to metal complexes confirmed the metal-ligand interaction, which is further supported by conductivity. The structural characteristics of organytellurium(IV) complexes were described using elemental analysis, IR, UV-vis, NMR and DFT.

**Molar conductivity:** In DMSO at 25 °C, the molar conductivity of all organytellurium complexes was calculated at approximately 10<sup>-3</sup> M. Table-1 shows the molar conductivity [32-34] values in ohm<sup>-1</sup> cm<sup>2</sup> mol<sup>-1</sup> of all organytellurium(IV) complexes (**2a-g**). This indicates that the metal complexes are weak-electrolytes.

TABLE-1  
MOLAR CONDUCTIVITY VALUES OF  
ORGANYLTELURUM(IV) COMPLEXES

Compound	Molar conductance (Ohm <sup>-1</sup> cm <sup>2</sup> mol <sup>-1</sup> )
<b>2a</b>	43.08
<b>2b</b>	33.25
<b>2c</b>	37.63
<b>2d</b>	43.93
<b>2e</b>	27.15
<b>2f</b>	32.13
<b>2g</b>	31.12

**Mass spectra:** The mass spectral data of a newly synthesized ligand as well as its organytellurium(IV) complexes were obtained. The molecular ion peak at *m/z* = 201.24 corresponds to the molecular mass of the ligand (*i.e.* molar mass = 201.22 g mol<sup>-1</sup>) as shown in the mass spectrum of Schiff base (HMeFPT). The molecular ion peak appear in the mass spectrum of various organytellurium(IV) complexes (**2a-g**) at *m/z* = 528.30, 542.31, 542.31, 585.93, 613.99, 613.99 and 436.89, which is identical to the molecular mass of the organytellurium(IV) complexes. According to mass analysis, RTeCl<sub>3</sub>·HMeFPT, R<sub>2</sub>TeCl<sub>2</sub>·HMeFPT and TeCl<sub>4</sub>·HMeFPT complexes have a (1:1) molar ratio.

**IR spectra:** Table-2 lists the major bands in the IR spectra of Schiff base ligand (L) and its organytellurium(IV) complexes [35-41]. The coordination sites were determined by comparing the IR spectra of complexes to that of free ligand. The O-H stretching frequency of the furanyl OH correlated to the occurrence of band in the area 3450-3150 cm<sup>-1</sup> in the spectra of ligand, HMeFPT. However, the position of this OH group in the complexes remained unaltered, demonstrating that -OH group of furan were not engaged in metal ion coordination. The C=N stretching frequency of free ligand occurs at 1627 cm<sup>-1</sup>. This band was moved to lower wavenumber [42] in complexes, showing that azomethine nitrogen was involved in the coordination to metal ion. In the ligand, a sharp intensity band owing to the (C-O-C) stretching vibration of furan emerged at 1287 cm<sup>-1</sup>. This band moved to a lower frequency in all the complexes, respectively, indicating that the furan

TABLE-2  
FT-IR-FREQUENCIES (cm<sup>-1</sup>) OF SCHIFF BASE (HMeFPT) AND ITS TELLURIUM(IV) COMPLEXES

Compound	v(O-H)	v(C-H) stretching	v(C-H) aromatic stretching	v(C-O-C)	v(C=N)	v(Te-O)	v(Te-N)
HMeFPT	-	2930 w	3057 m	1287 s	1634 s	-	-
<b>2a</b>	3348 b	2936 w	3064 w	1269 s	1618 s	289 m	507 m
<b>2b</b>	-	2939 w	3051 w	1273 s	1627 s	287 m	511 m
<b>2c</b>	3328 b	2931 w	3059 w	1267 s	1608 s	287 m	505 m
<b>2d</b>	3351 b	2937 w	3061 w	1231 s	1623 s	292 m	513 m
<b>2e</b>	-	2938 w	3072 w	1259 s	1598 s	288 m	507 m
<b>2f</b>	3361 b	2927 w	3068 w	1224 s	1627 s	290 m	510 m
<b>2g</b>	-	2935 w	3073 w	1243 s	1626 s	291 m	506 m



moiety is coordinated by oxygen. The  $\nu(\text{M-N})$  and  $\nu(\text{M-O})$  stretching vibrations were ascribed to the appearance of new bands in the spectra of organyltellurium(IV) [43-45] complexes in the ranges 513-505  $\text{cm}^{-1}$  and 292-286  $\text{cm}^{-1}$ , confirming that furanyl oxygen and azomethine *i.e.* nitrogen are implicated in the coordination. The data indicates that Schiff base (HMeFPT) ligand behave as a neutral bidentate coordinating through O donor and N donor atoms in complexes with distorted octahedral stereochemistry.

**$^1\text{H}$  and  $^{13}\text{C}$  NMR spectral studies:** The ligand as well as all the synthesized organyltellurium(IV) complexes have their  $^1\text{H}$  NMR spectra recorded in  $\text{DMSO-}d_6$  and thus their respective data is shown in Table-3. The  $^1\text{H}$  NMR spectra of the Schiff base ligand and their organyltellurium(IV) compounds gave additional evidence of the bonding mode of ligand. The parent ligand  $^1\text{H}$  NMR spectra revealed a singlet at  $\delta$  8.51 ppm, was ascribed to the azomethine ( $\text{CH}=\text{N}$ ), after complexation this azomethine signal showed deshielding and appear at in the range  $\delta$  8.56-8.97 ppm, implying coordination *via* the azomethine nitrogen atom of the ligand. Signal obtained in the synthesized ligand at  $\delta$  9.76 ppm ascribed to OH group remain unaffected in the spectra of complexes. Multiplets in the range  $\delta$  6.24-7.80 ppm were found in the ligand were

assigned to aromatic and furan ring protons after complexation the aromatic and furan ring protons were deshielded to region  $\delta$  6.24-7.91 ppm might be due to a decrease in electron density indicating that the azomethine nitrogen and furanyl oxygen atom are involved in coordination with tellurium metal [24-26,44]. Other singlet peak in the range of  $\delta$  2.49-2.55 ppm was assigned to  $\text{CH}_3$  indicating the presence of methyl group in the complexes. Table-4 lists the  $^{13}\text{C}$  NMR spectral data of HMeFPT ligand and its organyltellurium(IV) complexes. The  $^{13}\text{C}$  NMR spectra of ligand showed peak at 146.39 ppm is ascribed to azomethine carbon atom ( $\text{C}=\text{N}$ ) and shifted in the range of 163.76-167.81 ppm confirm their coordination to metal. The carbon atom attached to furanyl oxygen are identified at  $\delta$  139.49 and 124.61 ppm, respectively in the  $^{13}\text{C}$  NMR spectrum of Schiff base (HMeFPT) ligand shifted to downfield frequency [7] across all the studied organyltellurium(IV) complexes and were in the range 140-150 ppm implying coordination of ligand to tellurium metal *via* furanyl oxygen [46,47]. Additional signals were detected at  $\delta$  26.54 ppm, which were attributed to the carbon atoms of the  $\text{CH}_3$  group respectively.

**UV-Vis spectral studies:** UV-Vis spectra data have also been used to deduce the nature of the ligand field over the

TABLE-3  
 $^1\text{H}$  NMR SPECTRAL STUDIES OF SCHIFF BASE (HMeFPT) AND ITS Te(V) COMPLEXES

Compound	Chemical shift, $\delta$ ppm (in $\text{DMSO-}d_6$ )			
	Aromatic protons	OH proton	Methyl protons (4-toluidine ring)	$\text{CH}=\text{N}$ proton
HMeFPT	6.24-7.80	–	2.52	8.51
<b>2a</b>	6.27-7.84	10.57	2.53	8.67
<b>2b</b>	6.80-7.69	–	2.50	8.97
<b>2c</b>	6.91-7.88	9.43	2.51	8.61
<b>2d</b>	6.76-7.89	9.61	2.49	8.56
<b>2e</b>	6.85-7.91	–	2.55	8.83
<b>2f</b>	6.26-7.68	9.87	2.52	8.62
<b>2g</b>	6.53-7.79	–	2.47	8.73

TABLE-4  
 $^{13}\text{C}$  NMR SPECTRAL DATA OF SCHIFF BASE (HMeFPT) LIGAND AND ITS ORGANYLTELURUM(IV) COMPLEXES

Compound	Chemical shift, $\delta$ ppm ( $\text{DMSO-}d_6$ )		
	$\text{CH}=\text{N}$	C-H	Aromatic carbons
HMeFPT	146.39 (C6)	26.54 (C13)	139.49 (C2), 115.97 (C3), 115.42 (C4), 124.61 (C5), 131.44 (C7), 133.24 (C8, 12), 132.15 (C9, 11), 131.62 (C10),
<b>2a</b>	165.42 (C6)	22.51 (C13)	145.36 (C2), 109.48 (C3), 110.63 (C4), 145.28 (C5), 159.32 (C7), 124.36 (C8, C12), 136.26 (C9, C11), 136.92 (C10), 141.54 (C14), 139.41 (C15, 19), 137.46 (C16, 18), 153.28 (C17)
<b>2b</b>	167.23 (C6)	21.63 (C13), 54.79 (C20)	143.56 (C2), 111.52 (C3), 109.88 (C4), 146.51 (C5), 161.04 (C7), 123.98 (C8, C12), 137.79 (C9, C11), 139.62 (C10), 143.29 (C14), 140.35 (C15, 19), 137.25 (C16, 18), 153.09 (C17)
<b>2c</b>	167.34 (C6)	22.14 (C13), 54.97 (C20)	146.35 (C2), 111.52 (C3), 109.24 (C4), 148.35 (C5), 158.25 (C7), 125.63 (C8, C12), 140.53 (C9, C11), 138.96 (C10), 144.62 (C14), 141.73 (C15, 19), 135.40 (C16, 18), 157.89 (C17)
<b>2d</b>	165.39 (C6)	24.68 (C13)	158.03 (C2), 112.28 (C3), 110.79 (C4), 153.36 (C5), 143.71 (C7), 124.58 (C8, C12), 127.42 (C9, C11), 133.78 (C10), 129.46 (C14), 134.98 (C15, 19), 124.14 (C16, 18), 154.24 (C17), 127.46 (C20), 135.27 (C21, C25), 124.91 (C22, C24), 153.72 (C23)
<b>2e</b>	163.76 (C6)	25.03 (C13), 55.16 (C20), 54.87 (C27)	153.85 (C2), 111.38 (C3), 109.25 (C4), 143.71 (C5), 145.21 (C7), 123.58 (C8, C12), 126.14 (C9, C11), 132.83 (C10), 127.58 (C14), 133.41 (C15, 19), 128.35 (C16, 18), 140.61 (C17), 124.29 (C21), 134.52 (C22, C26), 129.83 (C23, C25), 140.34 (C24)
<b>2f</b>	167.81 (C6)	25.71 (C13), 55.24 (C20), 54.58 (C27)	147.51 (C2), 108.86 (C3), 109.32 (C4), 142.46 (C5), 143.52 (C7), 124.47 (C8, C12), 127.18 (C9, C11), 131.26 (C10), 128.67 (C14), 132.48 (C15, 19), 127.85 (C16, 18), 139.76 (C17), 124.72 (C21), 132.62 (C22, C26), 128.32 (C23, C25), 139.26 (C24),
<b>2g</b>	165.29 (C6)	25.61 (C13)	146.73 (C2), 106.81 (C3), 106.27 (C4), 142.81 (C5), 134.14 (C7), 132.48 (C8, C12), 131.68 (C9, C11), 132.62 (C10)

metal ion and the geometry of the complexes. Table-5 shows the UV-Vis spectral data of the ligand and its organytellurium (IV) complexes in solid state. In Fig. 1, UV-Vis spectra of the ligand (HMeFPT) revealed three bands at 224, 344 and 452 nm. The first band assigned to the  $\pi$ - $\pi^*$  [48] transition within the aromatic ring. This band displayed a bathochromic shift after complexation and was identified in the range 228-240 nm. The second band owing to the  $n$ - $\pi^*$  [49,50] transition due to the  $-C=N$  group shift to a longer wavelength in complexes and absorbs in the range 348-366 nm. The third band is attributed to the charge transfer within the furan moiety. Shift in wavelength in organytellurium(IV) complexes indicates the coordination of azomethine nitrogen and oxygen atom [23] to the tellurium metal.

Compound	$\lambda_{\max}$ ( $\pi$ - $\pi^*$ )	$\lambda_{\max}$ ( $n$ - $\pi^*$ )	$\lambda_{\max}$ (CT)
HMeFPT	224	344	452
<b>2a</b>	240	348	463
<b>2b</b>	228	366	463
<b>2c</b>	238	352	461
<b>2d</b>	229	356	463
<b>2e</b>	238	353	461
<b>2f</b>	239	349	462
<b>2g</b>	236	357	458

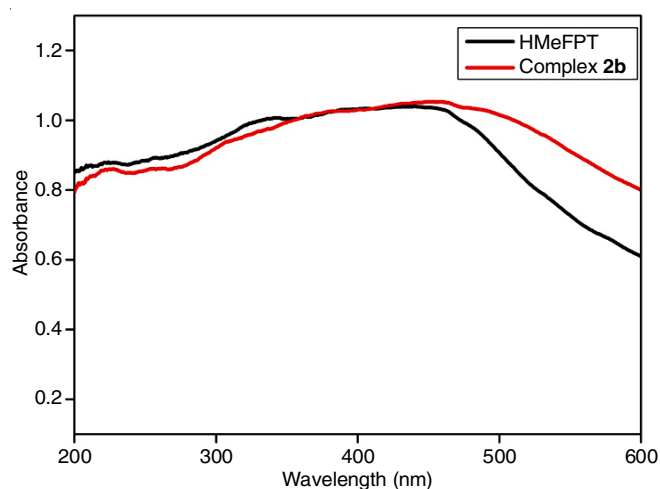


Fig. 1. UV-Vis spectra of the HMeFPT and complex (2b)

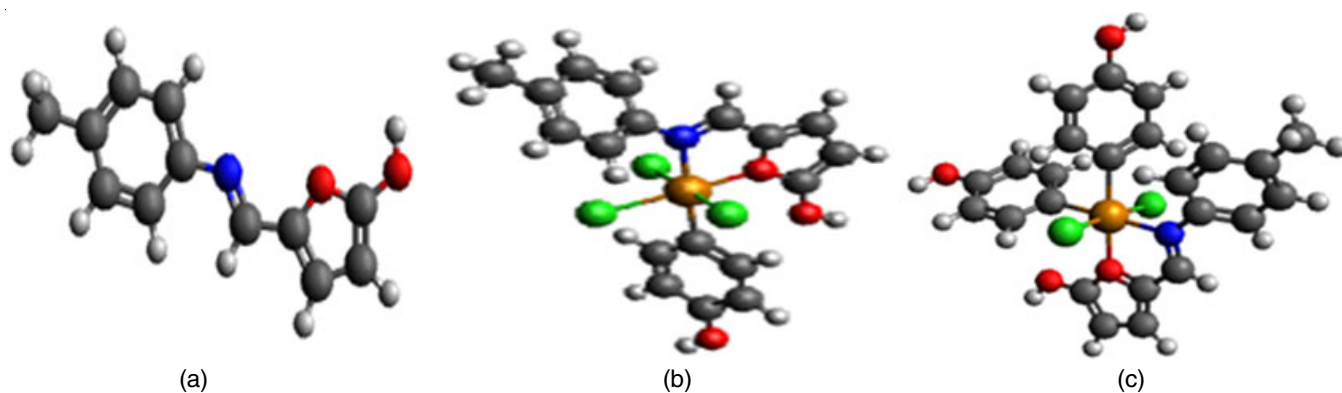


Fig. 2. Optimized structure of (a) HMeFPT (b) **2a** (c) **2d**. Colour code: blue-N; red-O; grey-C; white-H; green-Cl; yellow-Te

**Molecular modelling:** The 3D geometry of Schiff base (HMeFPT) as well as its organytellurium(IV) complexes (**2a** and **2d**) was optimized and DFT computations have been performed using the Orca program with the Avogadro 4.0 software. Fig. 2 shows the fully optimized molecular structures of the HMeFPT ligand and organytellurium(IV) complexes (**2a** and **2d**). The evaluated bond lengths are given in Table-6. The bond length and bond angle values around the tellurium metal are representative of their distorted octahedral geometry which is compatible with the hypothesized structures. The C-O-C and C=N (azomethine) bond lengths in the ligand are 1.315, 1.313 and 1.289, respectively. The C=N bond length get somewhat longer in complexes, whereas C-O-C bond length become slightly shorter. This alteration suggests that the azo-methine and C-O-C groups are linked *via* the N and O atoms. The distorted coordination sphere of metal is completed by three Te-Cl (2.41), Te-O (furanlyl) (2.024), Te-C (2.137) and Te-N (azomethine) (2.115).

Compound	C=N	C-O-C	Te-N (azomethine)	Te-O (furanlyl)
HMeFPT	1.289	1.315, 1.313	–	–
<b>2a</b>	1.298	1.294, 1.310	2.115	2.025
<b>2d</b>	1.299	1.295, 1.311	2.112	2.024

**DFT studies:** The essential molecular orbitals important for the chemical stability of compound are the highest occupied molecular orbitals (HOMO) and lowest unoccupied molecular orbitals (LUMO). For HMeFPT ligand and its organytellurium (IV) complexes **2a** and **2d**, DFT computations (def2-SVP) using Avogadro 4.0 and ORCA software provide HOMO and LUMO. The HOMO-LUMO energy gap (E) is an essential stability factor; the energy gap for Schiff base was found to be 5.155 eV, but the energy gap for complexes **2a** and **2d** was found to be 5.654 and 4.591 eV, respectively, due to the formation of a band in between the ligand and tellurium metal. The reactivity of the complexes increases due to decrease in lower energy gap. Quantum chemical parameters of the compounds, such like chemical potential ( $\mu$ ), hardness ( $\eta$ ), softness (S), electrophilicity index ( $\omega$ ) and electronegativity ( $\chi$ ) are calculated using HOMO and LUMO energies (Table-7). HOMO and LUMO are dispersed across the whole moiety in ligand. In complexes, LUMO is distributed throughout the whole moiety of the Schiff

TABLE-7  
THEORETICAL CALCULATED QUANTUM  
PARAMETERS OF HMeFPT, **2a** AND **2d**

Parameters	Ligand	<b>2a</b>	<b>2d</b>
$E_{\text{HOMO}}$ (eV)	-7.262	-6.117	-5.375
$E_{\text{LUMO}}$ (eV)	-2.107	-0.463	-0.784
$\Delta E$ (eV)	5.155	5.654	4.591
IE (eV)	7.262	6.117	5.375
$\chi$ (eV)	4.684	3.290	3.079
$\eta$ (eV)	2.577	2.827	2.295
S (eV) <sup>-1</sup>	0.194	0.176	0.217
$\omega$ (eV)	4.256	1.914	2.064
Pi	-4.684	-3.290	-3.079

base with little influence of tellurium metal whereas HOMO is distributed around the tellurium(IV) metal (Fig. 3).

**In vitro antimicrobial activity:** Antimicrobial activity of Schiff base (HMeFPT) and its organytellurium complexes (**2a-g**) were investigated against fungal strains (*C. albicans*, *A. niger*, *A. clavatus*) as well as Gram-positive and Gram-negative bacteria (*E. coli*, *P. aeruginosa*, *S. aureus*, *S. pyogenes*). Table-8 demon-

strated the minimum inhibition concentration values for the ligand and its metal complexes. Standard antibiotics such as ampicillin and greseofulvin were used. The results suggested that Schiff base ligand possess good antibacterial activity than antifungal activity. The synthesized organytellurium(IV) complexes are much effective than the free ligand by Overtone's concept and chelation theory. Organytellurium (IV) complexes such as **2a**, **2d** and **2f** have showed good antibacterial activity against *S. aureus*, while compounds **2c**, **2d** and **2f** possess better antibacterial activity against *E. coli*. Complex **2d** is the most active against all the studied bacterial strains. Complexes **2d**, **2e** and **2g** show excellent antifungal activity against *C. albicans*. Overall, complex **2e** is the most active against all fungal strains.

### Conclusion

5-((*p*-Tolylimino)methyl)furan-2-ol when reacted with tellurium tetrachloride, organytellurium chlorides (trichlorides and dichlorides) form 1:1 type complexes. Elemental analyses, conductance measurements, FT-Infrared, mass, UV-Vis, <sup>1</sup>H

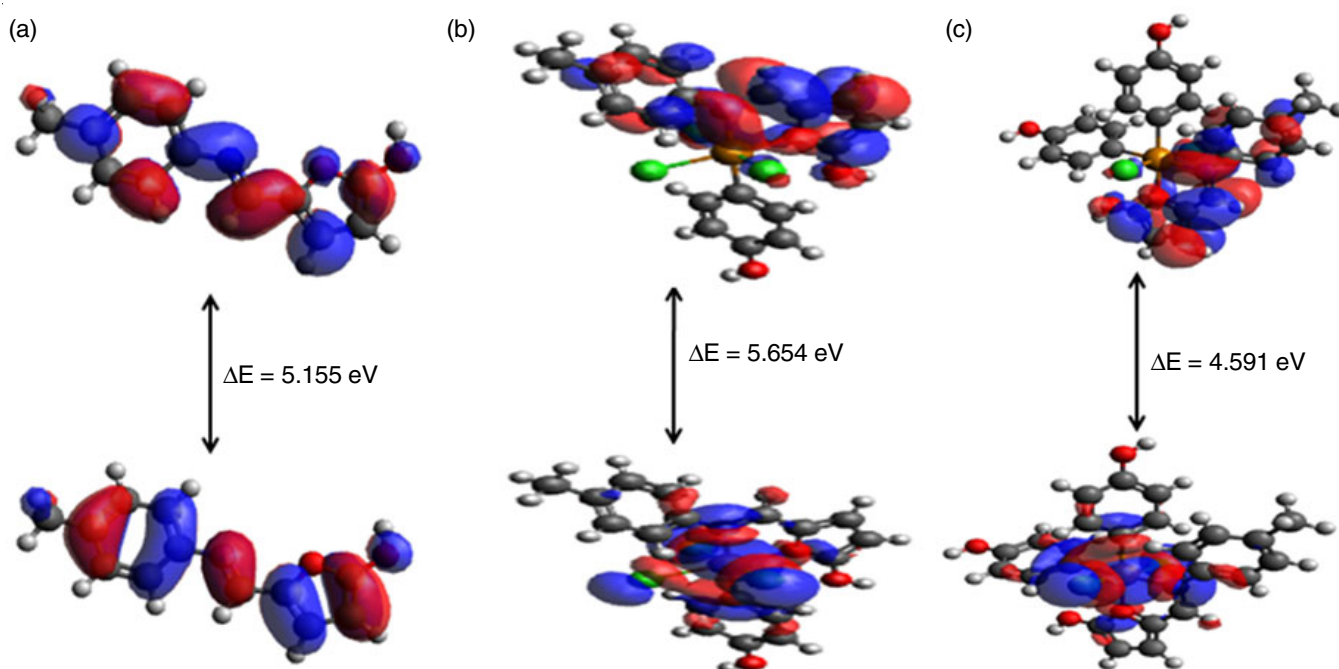


Fig. 3. HOMO and LUMO with energy gap of (a) HMeFPT (b) **2a** (c) **2d**

TABLE-8  
ANTIMICROBIAL ACTIVITIES OF SCHIFF BASE HMeFPT LIGAND AND ITS Te(IV) COMPLEXES

Compounds	Antibacterial activities				Antifungal activities		
	<i>S. aureus</i>	<i>S. pyogenes</i>	<i>E. coli</i>	<i>P. aeruginosa</i>	<i>C. albicans</i>	<i>A. niger</i>	<i>A. clavatus</i>
HMeFPT	0.20	0.1000	0.5000	0.2000	0.2	0.250	0.50
<b>2a</b>	0.20	0.2000	0.5000	0.2000	0.5	0.250	0.25
<b>2b</b>	0.50	0.5000	0.1250	0.5000	0.5	0.500	1.00
<b>2c</b>	0.25	0.1250	0.0625	1.0000	0.5	0.100	0.25
<b>2d</b>	0.10	0.0265	0.1000	0.0625	0.1	0.250	0.25
<b>2e</b>	0.50	0.2000	0.5000	0.1000	0.2	0.100	0.10
<b>2f</b>	0.20	0.5000	0.6250	0.2000	0.5	0.500	0.10
<b>2g</b>	0.50	0.1250	0.5000	0.2000	0.1	0.125	0.25
Ampicillin	0.25	0.1000	0.1	0.1000	–	–	–
Greseofulvin	–	–	–	–	0.5	0.100	0.10

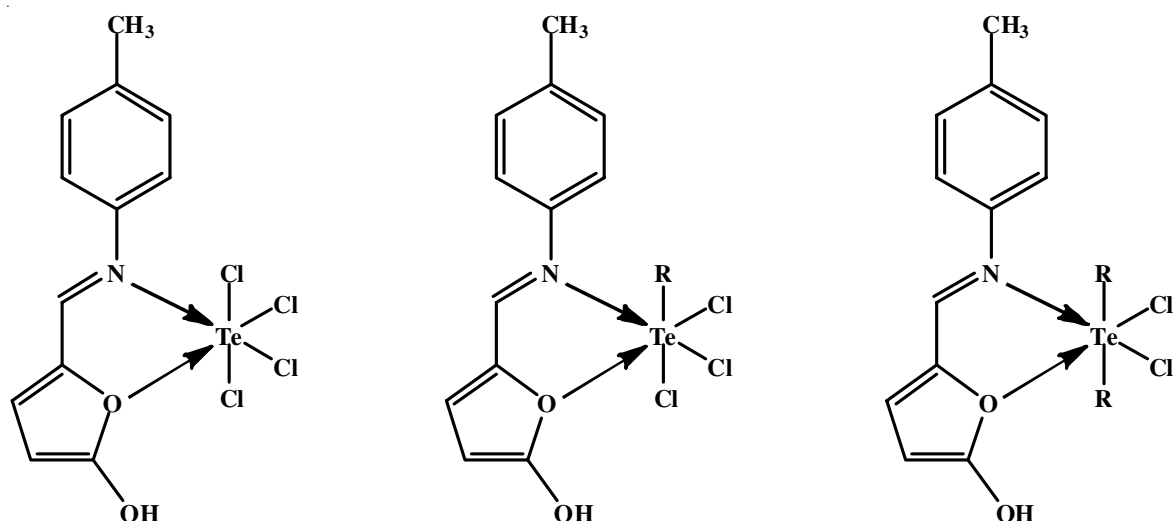


Fig. 4. Proposed structures of  $\text{TeCl}_4 \cdot \text{HMeFPT}$ ,  $\text{RTeCl}_3 \cdot \text{HMeFPT}$  and  $\text{R}_2\text{TeCl}_2 \cdot \text{HMeFPT}$

NMR and  $^{13}\text{C}$  NMR spectral investigations predict the neutral NO bidentate nature of ligand resulting in a distorted octahedral coordination around central tellurium atom. The geometry of the Schiff base (HMeFPT) and its organytellurium complexes has been optimized using molecular modelling. The Schiff base ligand and its organytellurium(IV) complexes were tested for antibacterial activity against bacterial and fungal species. According to the study, the synthesized organytellurium(IV) complexes have higher biological activity than the conventional drugs. The structures of these complexes have also been proposed in Fig. 4.

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#### CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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