

## Studies on Some Salicylhydroxamate Complexes of Aryltellurium(IV)

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**Abstract:** Twelve new salicylhydroxamate complexes of the type  $\text{ArTeCl}_2\cdot\text{SHA}$ ,  $\text{ArTeCl}\cdot(\text{SHA})_2$ ,  $\text{Ar}_2\text{TeCl}\cdot\text{SHA}$  and  $\text{Ar}_2\text{Te}\cdot(\text{SHA})_2$  (where Ar = *p*-methoxyphenyl, *p*-hydroxyphenyl, 3-methyl-4-hydroxyphenyl; SHA = salicylhydroxamate) have been synthesized by reactions of aryltellurium(IV) trichlorides and diaryltellurium(IV) dichlorides with potassium salicylhydroxamate. The aryltellurium(IV) hydroxamates have been characterized on the basis of elemental analyses, conductance measurements in DMSO, infrared and proton magnetic resonance studies. The spectral studies conjointly predicts the bidentate (*O*, *O'*) nature of hydroxamates to yield the penta and hexa-coordinated tellurium complexes. The biological activities of these complexes were evaluated against some pathogenic bacteria and fungi organisms.

**Keywords:** Salicylhydroxamate, Aryltellurium(IV), Diaryltellurium(IV), Antibacterial, Antifungal activities

### Introduction

The hydroxamic acids, with general formula  $\text{RCON}(\text{H})\text{OH}$ , are well known class of compounds due to their strong chelating properties. Hence they are involved in many biochemical processes such as iron transport phenomena<sup>1</sup>, inhibition of enzymatic activity of urease<sup>2,3</sup>, Alzheimer's Amyloid precursor protein  $\alpha$ -secretase<sup>4</sup> and some matrix metalloproteinases<sup>5</sup>. They also display antibacterial, anti-inflammatory and anti-asthmatic behaviour<sup>6,7</sup> and have been utilized in the design of therapeutic targets for cancer<sup>8,9</sup>, Alzheimer's disease<sup>10</sup>, haemochromatosis<sup>11,12</sup> and malaria<sup>13</sup>. Hydroxamic acids have been extensively studied as bioligands forming chelate complexes with numerous metals<sup>14-17</sup> including organotin complexes<sup>18,19</sup>.

Salicylhydroxamate ( $\text{SHA}^-$ ) can chelate *via* a number of possible coordination modes, not only the *O*, *O'*- mode, but also *N O'*- with the phenolato oxygen as *O*- donor, while most instance the hydroxamates are bound *via O*, *O'*- chelating<sup>20-27</sup>.

Also, aryltellurium(IV) trichlorides are known<sup>28-41</sup> to behave as Lewis acids and form complexes with several N-, O- and S- donor bases. The, diaryltellurium(IV) dichlorides are

also reported to act as acceptors but much weaker than aryltellurium(IV) trichlorides<sup>42-44</sup>. In view of this, it was thought desirable to study the reactions of potassium salicylhydroxamate with some aryltellurium trichlorides and diaryltellurium dichlorides. In this paper, we report the synthesis, characterization and antimicrobial studies on some new hydroxamates of the type  $\text{ArTeCl}_2\cdot\text{SHA}$ ,  $\text{ArTeCl}(\text{SHA})_2$ ,  $\text{Ar}_2\text{TeCl}\cdot\text{SHA}$  and  $\text{Ar}_2\text{Te}(\text{SHA})_2$  where, Ar = *p*-methoxyphenyl, *p*-hydroxyphenyl and 3-methyl-4-hydroxyphenyl and SHA = salicylhydroxamate.

## Experimental

All preparations were carried out under an atmosphere of dry nitrogen and the solvents used were purified by standard method<sup>45,46</sup> before use. The purity of compounds was checked by TLC using Silica gel-G (Merck). Melting points were determined in open capillary tube and are uncorrected.

Carbon, hydrogen and nitrogen analyses were obtained microanalytically from SAIF, Panjab University Chandigarh on a ThermoFinnigan CHNS analyser. Conductivity was measured in DMSO at  $25\pm 2$  °C with a microprocessor based conductivity bridge type MICROSIL.

IR spectra were recorded in KBr pellets at SAIF, Panjab University Chandigarh on a F.T. Infra-Red spectrophotometer Model RZX (Perkin Elmer). <sup>1</sup>H NMR spectra were recorded in DMSO-*d*<sub>6</sub> using TMS as an internal reference on BRUKER AVANCE II 400 NMR spectrometer. The antimicrobial screening was carried out by tube dilution method at Department of Pharmaceutical Sciences, M. D. University, Rohtak.

### *Preparation of aryltellurium(IV) trichlorides and diaryltellurium(IV) dichlorides*

*p*-Methoxyphenyltellurium(IV)trichloride<sup>47,48</sup>, bis(*p*-methoxyphenyl)tellurium(IV) dichloride<sup>48,49</sup>, *p*-hydroxyphenyltellurium(IV) trichloride<sup>50</sup>, bis(*p*-hydroxyphenyl) tellurium (IV) dichloride<sup>50</sup>, 3-methyl-4-hydroxyphenyltellurium(IV) trichloride<sup>51</sup> and bis(3-methyl-4-hydroxyphenyl)-tellurium(IV) dichloride<sup>51</sup> were prepared by the reactions of tellurium tetrachloride with corresponding arenes *i.e.* anisole, phenol, *o*-cresol respectively, by the methods reported in the literature.

### *Preparation of potassium salicylhydroxamate (KSHA)*

Potassium salicylhydroxamate was prepared in two steps as follows:

#### *Preparation of ethyl ester of salicylic acid*<sup>45</sup>

To 0.25 mole of salicylic acid, excess (up to 2.5 moles) of ethyl alcohol and 1 mL of conc.  $\text{H}_2\text{SO}_4$  in separate reaction flasks were added. The reactants were refluxed for about 3 h till whole of the acid dissolved in ethanol. The contents were cooled and transferred to 60 mL of water in a separating funnel. The lower layer of ester was removed and was washed with saturated sodium bicarbonate until the effervescence ceased. Finally washed with water and dried over anhydrous sodium sulphate.

#### *Preparation of potassium salt of hydroxamic acid*

The potassium salt was prepared by the method reported by Houser *et al.*,<sup>52</sup>. Cooled solution of KOH (28.05 g in 70 mL methanol) was added to methanolic solution of hydroxylamine (23.27 g in 120 mL) with constant shaking and cooling. The mixture was allowed to cool for 24 hours in an ice bath to ensure complete precipitation of KCl, which was removed by filtration. To this filtrate was added 25 mL of ethyl ester of salicylhydroxamic acid. The reaction mixture was kept in air tight flask at room temperature for 2-3 days to yield the fine crystals of potassium salicylhydroxamate, which were filtered and dried in air. Yield 80%, m. pt. 260-270 °C.

### Preparation of aryltellurium(IV) salicylhydroxamates

Aryltellurium(IV) trichlorides,  $\text{ArTeCl}_3$  (Ar = *p*-methoxyphenyl, *p*-hydroxyphenyl, 3-methyl-4-hydroxyphenyl), when treated with potassium salicylhydroxamate in different molar ratios, yield  $\text{ArTeCl}_2\cdot\text{SHA}$  and  $\text{ArTeCl}\cdot(\text{SHA})_2$  type complexes.

#### $\text{ArTeCl}_2\cdot\text{SHA}$

A warm saturated methanolic solution of potassium salicylhydroxamate (0.38 g, 2 mmol) was added dropwise to a solution of aryltellurium(IV) trichloride (2 mmol) in chloroform/methanol. An immediate precipitation of KCl resulted which was removed by filtration. The filtrate was further refluxed for 3-4 hours to precipitate out any KCl and clear solution was then concentrated to about one third of original volume and kept overnight to yield crystalline product. This was filtered, washed with chloroform and dried in a vacuum desiccators over  $\text{P}_4\text{O}_{10}$ .

#### $\text{ArTeCl}\cdot(\text{SHA})_2$

The saturated solution of aryltellurium(IV) trichloride (2 mmol) in chloroform/methanol was added dropwise with constant stirring to a saturated methanolic solution of potassium salicylhydroxamate (0.76 g, 4 mmol). An immediate change in colour with precipitation of KCl took place, which was removed by filtration. The contents were refluxed for 3-4 hours and then clear solution concentrated to about one third of original volume and left overnight to get coloured crystalline product, which was filtered, washed with chloroform and dried in a vacuum desiccators over  $\text{P}_4\text{O}_{10}$ .

### Preparation of diaryltellurium(IV) salicylhydroxamates

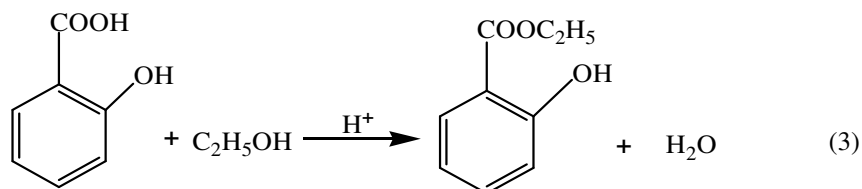
Diaryltellurium(IV) dichlorides,  $\text{Ar}_2\text{TeCl}_2$  (Ar = *p*-methoxyphenyl, *p*-hydroxyphenyl and 3-methyl-4-hydroxyphenyl), when treated with potassium salicylhydroxamate yield both 1:1 and 1:2 complexes of the type  $\text{Ar}_2\text{TeCl}\cdot\text{SHA}$  and  $\text{Ar}_2\text{Te}\cdot(\text{SHA})_2$ . These have been synthesized by the same procedure as for salicylhydroxamate of aryltellurium(IV) described above.

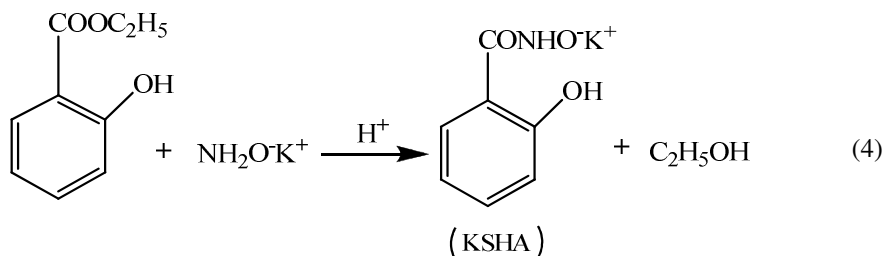
## Results and Discussion

Tellurium tetrachloride when heated with anisole<sup>47-49</sup>, phenol<sup>50</sup>, *o*-cresol<sup>51</sup> (Ar-H) appears to undergo Friedel-Crafts type condensation reaction whereby  $\text{TeCl}_3^+$  unit attacks a position *para* to the  $-\text{OH}/-\text{OCH}_3$  groups in the aromatic rings, thus resulting in the formation of aryltellurium(IV) trichlorides and diaryltellurium(IV) dichlorides.

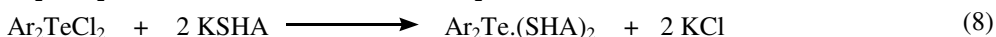


Preparation of potassium salicylhydroxamate involves in two steps: first the preparation of ethyl ester of salicylic acid and then the potassium salt, which can be represented by following equations.





Potassium salicylhydroxamate (KSHA) reacts with aryltellurium(IV) trichlorides and diaryltellurium(IV) dichlorides in 1:1 and 2:1 molar ratios to yield the corresponding aryltellurium(IV) hydroxamate.



These new aryltellurium(IV) hydroxamates have been analysed for tellurium, chloride, carbon, hydrogen and nitrogen contents and the data along with their physical properties and yields are presented in Table 1.

### Conductance studies

Molar conductance ( $\Lambda_M$ ) data for aryltellurium(IV) hydroxamates in DMSO are compiled in Table 1. The  $\Lambda_M$  values at *ca.*  $10^{-3}$  M predicts the weak to 1:1 electrolyte<sup>53,54</sup> type behavior of these hydroxamates in DMSO, probably due to ionization into  $\text{ArTeCl}.\text{SHA}^+/\text{ArTe}.\text{(SHA)}_2^+/\text{Ar}_2\text{Te}.\text{SHA}^+$  and  $\text{Cl}^-$  in DMSO. The higher  $\Lambda_M$  values for  $\text{Ar}_2\text{Te}.\text{(SHA)}_2$  may be due to steric factors and donor behavior of DMSO to result in probable dissociation into  $\text{Ar}_2\text{Te}.\text{SHA}.\text{DMSO}^+$  and  $\text{SHA}^-$  ions.

### Infrared spectra

The infrared spectra of the aryltellurium(IV) hydroxamates are quite complex and an attempt has therefore been made to identify the donor sites of hydroxamate ligand by comparing with those of parent aryltellurium(IV) chlorides and potassium hydroxamate, which illustrated clear differences.

The principal infrared absorption bands of KSHA are those due to  $\nu_{(\text{C}=\text{O})}$ ,  $\nu_{(\text{C}-\text{N})}$ ,  $\nu_{(\text{N}-\text{O})}$  and  $\nu_{(\text{N}-\text{H})}$  stretching vibrations of the hydroxamate group observed in the spectrum at  $1606 \text{ cm}^{-1}$ ,  $1392 \text{ cm}^{-1}$ ,  $999 \text{ cm}^{-1}$  and  $3287\text{-}3233 \text{ cm}^{-1}$  respectively (Table 2).

The absorption band occurring at  $1606 \text{ cm}^{-1}$  in KSHA attributes to  $\nu_{(\text{C}=\text{O})}$  mode, shifted to lower wave numbers and appeared at  $1576\text{-}1588 \text{ cm}^{-1}$  in aryltellurium(IV) hydroxamates. The absorption band due to  $\nu_{(\text{C}-\text{N})}$  mode occurring at  $1392 \text{ cm}^{-1}$  in free KSHA has been found to shift towards higher region at  $1453\text{-}1456 \text{ cm}^{-1}$  in the complexes. The band at  $3233\text{-}3288 \text{ cm}^{-1}$  due to  $\nu_{(\text{N}-\text{H})}$  mode in KSHA did not undergo any change, however could not be ascertained due to phenolic OH group in the aryltellurium moiety. This rules out the involvement of coordination through nitrogen atom. The sharp band occurring at  $999 \text{ cm}^{-1}$  in KSHA ascribed to  $\nu_{(\text{N}-\text{O})}$  mode has been observed to move towards higher wave number and appeared at  $1025\text{-}1032 \text{ cm}^{-1}$  in aryltellurium(IV) salicylhydroxamates.

**Table 1.** Analytical data, molar conductance and physical properties of aryltellurium(IV) salicylhydroxamates

| Compd No. | Complex (Ar)  | Empirical Formula (Formula Wt.)  | Colour, Yield, %    | M. Pt., °C dec. | Analyses % Found (Calculated) |                |                |                  |                  | $\Lambda_M$ at ca. $10^{-3}$ M $\text{ohm}^{-1} \text{cm}^2 \text{mol}^{-1}$ in DMSO |
|-----------|---|--|---------------------|-----------------|-------------------------------|----------------|----------------|------------------|------------------|--|
|           |   |  |                     |                 | C                             | H              | N              | Te               | Cl               |  |
| I         | ArTeCl <sub>2</sub> .SHA<br>( <i>p</i> -methoxyphenyl)              | C <sub>14</sub> H <sub>13</sub> Cl <sub>2</sub> NO <sub>4</sub> Te<br>(457.66) | Dull white<br>(92)  | 75-78           | 36.44<br>(36.71)              | 2.72<br>(2.84) | 2.86<br>(3.06) | 28.70<br>(27.87) | 15.25<br>(15.51) | 37.63  |
| II        | ArTeCl.(SHA) <sub>2</sub><br>( <i>p</i> -methoxyphenyl)             | C <sub>21</sub> H <sub>19</sub> ClN <sub>2</sub> O <sub>7</sub> Te<br>(574.29) | Cream<br>(90)       | 85-90           | 43.51<br>(43.86)              | 3.12<br>(3.31) | 5.12<br>(4.87) | 21.92<br>(22.21) | 5.98<br>(6.17)   | 42.31  |
| III       | ArTeCl <sub>2</sub> .SHA<br>( <i>p</i> -hydroxyphenyl)              | C <sub>13</sub> H <sub>11</sub> Cl <sub>2</sub> NO <sub>4</sub> Te<br>(443.65) | Light grey<br>(94)  | 74-76           | 34.72<br>(35.14)              | 2.79<br>(2.48) | 3.35<br>(3.16) | 28.52<br>(28.76) | 15.74<br>(16.00) | 48.41  |
| IV        | ArTeCl.(SHA) <sub>2</sub><br>( <i>p</i> -hydroxyphenyl)             | C <sub>20</sub> H <sub>17</sub> ClN <sub>2</sub> O <sub>7</sub> Te<br>(560.28) | Light brown<br>(95) | 78-80           | 42.52<br>(42.82)              | 3.24<br>(3.03) | 4.98<br>(5.00) | 22.43<br>(22.77) | 6.40<br>(6.33)   | 25.22  |
| V         | ArTeCl <sub>2</sub> .SHA<br>(3-methyl-4-hydroxyphenyl)              | C <sub>14</sub> H <sub>13</sub> Cl <sub>2</sub> NO <sub>4</sub> Te<br>(457.66) | Brown<br>(85)       | 50-52           | 35.98<br>(36.71)              | 2.99<br>(2.84) | 2.98<br>(3.06) | 27.41<br>(27.87) | 14.98<br>(15.51) | 48.71  |
| VI        | ArTeCl.(SHA) <sub>2</sub><br>(3-methyl-4-hydroxyphenyl)             | C <sub>21</sub> H <sub>19</sub> ClN <sub>2</sub> O <sub>7</sub> Te<br>(574.29) | Pale yellow<br>(82) | 60-64           | 42.60<br>(43.86)              | 3.63<br>(3.31) | 4.93<br>(4.87) | 22.06<br>(22.21) | 6.42<br>(6.17)   | 39.15  |
| VII       | Ar <sub>2</sub> TeCl.SHA<br>( <i>p</i> -methoxyphenyl)              | C <sub>21</sub> H <sub>20</sub> ClNO <sub>5</sub> Te<br>(529.29)               | Cream<br>(84)       | 115-120         | 46.76<br>(47.59)              | 3.99<br>(3.78) | 2.30<br>(2.65) | 25.12<br>(24.10) | 7.37<br>(6.70)   | 25.83  |
| VIII      | Ar <sub>2</sub> Te.(SHA) <sub>2</sub><br>( <i>p</i> -methoxyphenyl) | C <sub>28</sub> H <sub>26</sub> N <sub>2</sub> O <sub>8</sub> Te<br>(645.92)   | Brown<br>(87)       | 110-112         | 52.87<br>(52.00)              | 4.43<br>(4.02) | 4.17<br>(4.33) | 19.07<br>(19.75) | -                | 73.12  |
| IX        | Ar <sub>2</sub> TeCl.SHA<br>( <i>p</i> -hydroxyphenyl)              | C <sub>19</sub> H <sub>16</sub> ClNO <sub>5</sub> Te<br>(501.26)               | Pink<br>(89)        | 100-105         | 44.71<br>(45.47)              | 3.46<br>(3.19) | 3.12<br>(2.79) | 26.39<br>(25.45) | 7.23<br>(7.08)   | 20.24  |
| X         | Ar <sub>2</sub> Te.(SHA) <sub>2</sub><br>( <i>p</i> -hydroxyphenyl) | C <sub>26</sub> H <sub>22</sub> N <sub>2</sub> O <sub>8</sub> Te<br>(617.89)   | Orange<br>(91)      | 86-88           | 49.99<br>(50.48)              | 3.98<br>(3.56) | 4.67<br>(4.53) | 21.01<br>(20.65) | -                | 22.88  |
| XI        | Ar <sub>2</sub> TeCl.SHA<br>(3-methyl-4-hydroxyphenyl)              | C <sub>21</sub> H <sub>20</sub> ClNO <sub>5</sub> Te<br>(529.29)               | Red<br>(78)         | 120-122         | 47.65<br>(47.59)              | 3.60<br>(3.78) | 2.12<br>(2.65) | 25.22<br>(24.10) | 6.36<br>(6.70)   | 11.64  |
| XII       | Ar <sub>2</sub> Te.(SHA) <sub>2</sub><br>(3-methyl-4-hydroxyphenyl) | C <sub>28</sub> H <sub>26</sub> N <sub>2</sub> O <sub>8</sub> Te<br>(645.92)   | Grey<br>(75)        | 148-150         | 52.36<br>(52.00)              | 4.49<br>(4.02) | 4.20<br>(4.33) | 18.93<br>(19.75) | -                | 71.19  |

Values of  $\Lambda_M$  reported<sup>53, 54</sup> for 1:1 electrolytes in DMSO = 50 – 70  $\text{ohm}^{-1} \text{cm}^2 \text{mol}^{-1}$

A shift in  $\nu_{(C=O)}$  mode to lower wave number and  $\nu_{(N-O)}$  mode to higher wave numbers are suggestive of bonding of salicylhydroxamate ion via oxygen atoms of carbonyl and hydroxylamine group<sup>19,23,55-58</sup>. The appearance of new weak bands around 283-297  $\text{cm}^{-1}$  due to  $\nu_{(Te-O)}$ <sup>59-61</sup> confirms the bonding of ligand to tellurium through the oxygen atoms. Also, independent assignment of  $\nu_{O-H}$  (phenolic) could not be made due to presence of this group in hydroxyaryltellurium(IV) moiety *i.e.*  $\text{ArTe}$  or  $\text{Ar}_2\text{Te}$ .

### <sup>1</sup>H NMR spectra

Proton magnetic resonance spectra of aryltellurium(IV) salicylhydroxamates are very complex and a lot of mixing of aryl proton singals of the salicylhydroxamate and aryltellurium(IV) moiety takes place, thus making the independent assignment almost impossible. The chemical shift data for the free salicylhydroxamic acid<sup>62</sup> and aryltellurium(IV) salicylhydroxamates are compiled in the Table 3.

**Table 2.** Important infrared absorption bands ( $\text{cm}^{-1}$ ) of potassium salicylhydroxamate and aryltellurium(IV) hydroxamates

| Compound | $\nu_{(C=O)}$ | $\nu_{(C-N)}$ | $\nu_{(N-O)}$ | $\nu_{(Te-O)}$ |
|----------|---------------|---------------|---------------|----------------|
| KSHA     | 1606 s        | 1392 vs       | 999 s         | -              |
| I        | 1580 s        | 1454 m        | 1029 m        | 288 w          |
| II       | 1579 s        | 1454 m        | 1032 m        | 297 w          |
| III      | 1577 s        | 1454 m        | 1032 m        | 289 w          |
| IV       | 1577 s        | 1454 m        | 1032 m        | 283 w          |
| V        | 1576 s        | 1453 m        | 1031 m        | 288 w          |
| VI       | 1577 s        | 1454 s        | 1032 m        | 288 w          |
| VII      | 1583 s        | 1454 s        | 1028 m        | 290 w          |
| VIII     | 1584 s        | 1456 m        | 1027 m        | 290 w          |
| IX       | 1577 vs       | 1453 s        | 1032 m        | 288 w          |
| X        | 1577 vs       | 1453 s        | 1032 m        | 288 w          |
| XI       | 1588 vs       | 1454 s        | 1025 m        | 292 w          |
| XII      | 1588 vs       | 1454 m        | 1026 m        | 290 w          |

*s* = strong, *vs* = very strong, *m* = medium, *w* = weak

**Table 3.** <sup>1</sup>H NMR spectral data of aryltellurium(IV) salicylhydroxamates in DMSO-d<sub>6</sub>

| Compound                                    | Chemical Shift, $\delta$ ppm  |
|---|---|
| $\text{C}_6\text{H}_4(\text{OH})$<br>CONHOH | 6.82(q, 1H), 6.86(d, 1H), 7.35(q, 1H), 7.65(d, 1H), 9.32(s, 1H, -NOH),<br>11.43(s, 1H, -NH), 12.20(s, 1H, phenolic OH)  |
| I   | 3.87(s, 3H, -OCH <sub>3</sub> ), 6.81- 8.38(cm, 8H, aryl protons of ArTe and SHA),<br>11.46(s, 1H, -NH), 12.40(bs, 1H, phenolic OH of SHA)                          |
| II  | 3.83(s, 3H, -OCH <sub>3</sub> ), 6.81- 8.39(cm, 12H, aryl protons of ArTe and SHA),<br>11.45(s, 2H, -NH), 12.30(bs, 2H, phenolic OH of SHA)                         |
| III   | 6.83-7.81(cm, 8H, aryl protons of ArTe and SHA), 8.24(s, 1H, OH of<br>ArTe), 11.45(s, 1H, -NH), 10.09(bs, 1H, phenolic OH of SHA)                                   |
| IV  | 6.817-7.94(cm, 12H, aryl protons of ArTe and SHA), 8.21(s, 1H, OH of<br>ArTe), 11.45(s, 2H, -NH), 12.264(bs, 2H, phenolic OH of SHA)                                |
| V   | 2.16(s, 3H, -CH <sub>3</sub> ), 6.74-7.83(cm, 7H, aryl protons of ArTe and SHA),<br>8.11(s, 1H, OH of ArTe), 11.45(s, 1H, -NH), 9.97(bs, 1H, phenolic OH of<br>SHA) |

Contd....

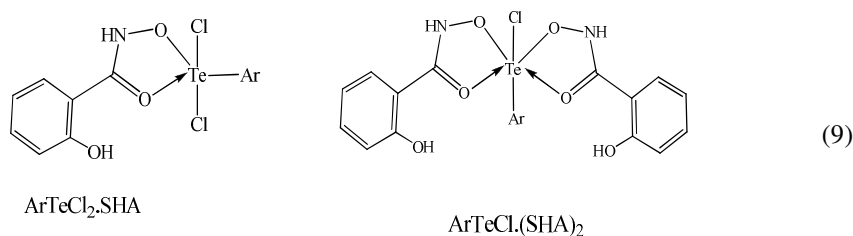
|      |   |
|------|---|
| VI   | 2.16(s, 3H, -CH <sub>3</sub> ), 6.82-7.81(cm, 11H, aryl protons of ArTe and SHA), 8.11(s, 1H, OH of ArTe), 11.457(s, 2H, -NH), 10.10, 12.31(bs, 2H, phenolic OH of SHA)           |
| VII  | 3.83(s, 6H, -OCH <sub>3</sub> ), 6.815- 8.008(cm, 12H, aryl protons of Ar <sub>2</sub> Te and SHA), 11.47(s, 1H, -NH), 12.296(s, 1H, phenolic OH of SHA)                          |
| VIII | 3.82(s, 6H, -OCH <sub>3</sub> ), 6.816- 7.977(cm, 16H, aryl protons of Ar <sub>2</sub> Te and SHA), 11.48(s, 2H, -NH), 12.29(s, 2H, phenolic OH of SHA)                           |
| IX   | 6.767-7.879(cm, 12H, aryl protons of Ar <sub>2</sub> Te and SHA), 9.33(s, 2H, OH of Ar <sub>2</sub> Te), 11.46(s, 1H, -NH), 12.286(s, 1H, phenolic OH of SHA)                     |
| X    | 6.774-7.828(cm, 16H, aryl protons of Ar <sub>2</sub> Te and SHA), 9.37(s, 2H, OH of Ar <sub>2</sub> Te), 11.467(s, 2H, -NH), 12.257(s, 2H, phenolic OH of SHA)                    |
| XI   | 2.53(s, -CH <sub>3</sub> ), 6.218-7.757(cm, aryl protons of Ar <sub>2</sub> Te and SHA), 8.20(s, OH of Ar <sub>2</sub> Te), poor solubility, spectra not well resolved            |
| XII  | 2.51(s, -CH <sub>3</sub> ), 6.628-7.749(cm, aryl protons of Ar <sub>2</sub> Te and SHA), 8.26(s, OH of Ar <sub>2</sub> Te), spectra not well resolved due to very poor solubility |

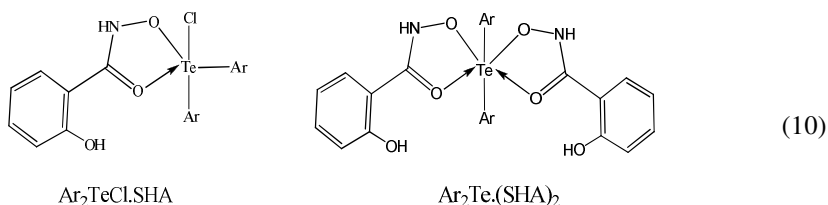
*s* = singlet, *d* = doublet, *q* = quartet, *cm* = complex multiplet, *bs* = broad singlet

Free salicylhydroxamic acid shows four separate singals due to aryl proton at 6.82, 6.86, 7.35 and 7.65  $\delta$  ppm. The hydroxamic N-H and O-H protons resonate at 11.43 and 9.32  $\delta$  ppm respectively, and phenolic OH at 12.20  $\delta$  ppm as singlets. The hydroxamic OH singlet at 9.32  $\delta$  ppm disappears in all the complexes indicating thereby the deprotonation of -NOH proton and subsequently linkage of tellurium with this oxygen atom. The N-H proton signal at  $\delta$  11.43 in free salicylhydroxamic acid, also is observed in all the salicylhydroxamate complexes, confirming its non participation in the bonding with tellurium atom. The broad singlet around 12 ppm indicates that phenolic OH of salicylhydroxamate is not involved in bonding with the tellurium. The spectra of complexes of bis(3-methyl-4-hydroxyphenyl)-tellurium(IV) are not well resolved due to very poor solubility in DMSO-d<sub>6</sub>.

Further, the aryl protons of aryltellurium(IV), diaryltellurium(IV) and salicylhydroxamate groups exhibit a lot of overlapping of signals and are observed as complex multiplet in the region 6.218-8.39  $\delta$  ppm, as observed in <sup>1</sup>H NMR Spectra of organotin(IV) complexes of hydroxamic acids<sup>19</sup>. Also, a careful examination of <sup>1</sup>H NMR Spectra of hydroxamate complexes reveal the shielding of aryl protons of ArTe/Ar<sub>2</sub>Te compared<sup>50,51,63,64</sup> to ArTeCl<sub>3</sub>/Ar<sub>2</sub>TeCl<sub>2</sub> and deshielding of protons of SHA due to flow of electron density from the ligand to the aryltellurium moiety as a result of complexation.

Thus, on the basis of infrared and proton magnetic resonance spectral studies it may be concluded that SHA<sup>-</sup> acts as a bidentate (*O*, *O*) ligand involving the hydroxamate (-NHO) and carbonyl oxygens, giving rise to penta coordinated tellurium complexes in ArTeCl<sub>2</sub>.SHA and Ar<sub>2</sub>TeCl<sub>2</sub>.SHA and hexa coordinated in ArTeCl<sub>2</sub>(SHA)<sub>2</sub> and Ar<sub>2</sub>Te<sub>2</sub>(SHA)<sub>2</sub>. The proposed structures (Figure 1) are as below:





**Figure 1.** Proposed structure of complexes

### Antimicrobial activity

All the newly synthesized complexes were investigated for their *in vitro* antimicrobial potential against gram positive bacteria (*S. aureus* and *B. subtilis*), gram negative bacterium (*E. coli*) and fungal strains (*C. albicans* and *A. niger*) by tube dilution method. The minimum inhibitory concentration (MIC) values are presented in the Table 4.

**Table 4.** Minimum inhibitory concentration of potassium salicylhydroxamate and aryltellurium(IV) hydroxamates.

| Compound | MIC, $\mu\text{g/mL}$ |                    |                |                 |                    |
|----------|-----------------------|--------------------|----------------|-----------------|--------------------|
|          | <i>S. aureus</i>      | <i>B. subtilis</i> | <i>E. coli</i> | <i>A. niger</i> | <i>C. albicans</i> |
| KSHA     | 12.5                  | 12.5               | 12.5           | 12.5            | 6.25               |
| I        | 12.5                  | 12.5               | 6.25           | 6.25            | 6.25               |
| II       | 25                    | 12.5               | 12.5           | 6.25            | 6.25               |
| III      | 12.5                  | 12.5               | 6.25           | 12.5            | 12.5               |
| IV       | 12.5                  | 25                 | 6.25           | 12.5            | 12.5               |
| V        | 12.5                  | 12.5               | 6.25           | 12.5            | 6.25               |
| VI       | 12.5                  | 12.5               | 6.25           | 6.25            | 6.25               |
| VII      | 12.5                  | 25                 | 6.25           | 6.25            | 6.25               |
| VIII     | 12.5                  | 12.5               | 12.5           | 6.25            | 6.25               |
| IX       | 12.5                  | 25                 | 12.5           | 6.25            | 6.25               |
| X        | 12.5                  | 25                 | 12.5           | 6.25            | 6.25               |
| XI       | 12.5                  | 12.5               | 12.5           | 6.25            | 6.25               |
| XII      | 12.5                  | 12.5               | 12.5           | 6.25            | 6.25               |

The MIC data reveal that the aryltellurium(IV) hydroxamate complexes and potassium hydroxamate possess more biocidal activity than the precursor salicylhydroxamic acid<sup>22</sup>, which shows MIC in the range 66-88  $\mu\text{g/mL}$ .

### Conclusion

Aryltellurium(IV) trichlorides and diaryltellurium(IV) dichlorides upon reaction with potassium salicylhydroxamate in different molar ratios yield the 1:1 and 1:2 type complexes, whereby chloride is exchanged with salicylhydroxamate group. The hydroxamate complexes have been characterized by elemental analyses, conductance measurement, infrared and proton magnetic resonance spectral studies. The salicylhydroxamate in these complexes functions as a bidentate ligand through carbonyl and hydroxamate oxygen atoms. The complexes have been observed to possess appreciable antimicrobial activity against bacteria and fungi.

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