



STUDIES ON SOME TELLURIUM (IV) COMPLEXES OF N, N'-BIS (INDOL-2-OXO-3-YLIDENE)- 1, 2-DIAMINOETHANE

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ABSTRACT

A series of new tellurium(IV) chelates with Schiff base formed by condensation of isatin with ethylenediamine having formulae $\text{TeCl}_4(\text{H}_2\text{L})$, $\text{RTeCl}_3(\text{H}_2\text{L})$ and $\text{R}_2\text{TeCl}_2(\text{H}_2\text{L})$; where R = *p*-methoxy-, *p*-hydroxy- and 3-methyl-4-hydroxyphenyl and H_2L = Schiff base, have been synthesized and characterized *via* elemental analyses, conductance measurement, infrared and proton magnetic resonance spectral studies. The data predict the hexacoordination of central tellurium atom by the neutral ONNO quadridentate Schiff base. Some of these complexes have also been observed to possess antifungal and antitubercular activity.

Key words: Ethylenediamine, Isatin, Schiff base, Tellurium (IV), Quadridentate, Antimicrobial activity.

INTRODUCTION

Isatin (Indole-2, 3-dione) and its derivatives are of interest due to their biological and medical applications such as anticonvulsant^{1,2}, anti-HIV³, cytotoxic⁴, tuberculostatic⁵, and antimicrobial⁶ activity. A large number of Schiff's bases of isatin have been reported in the literature, which undergoes complexation with various metal ions in different modes⁷⁻¹⁵.

Also, tellurium (IV) compounds such as TeCl_4 ¹⁶⁻¹⁸, aryltellurium (IV) trichlorides¹⁹⁻³² and diaryltellurium (IV) dichlorides³³⁻³⁵ are reported to act as acceptors and form complexes with nitrogen, oxygen and sulphur donor bases. In view of this and in continuation of earlier work on isatin Schiff bases³⁶, we herein report the studies on complexes of tellurium (IV) chloride, aryltellurium (IV) trichlorides, RTeCl_3 and diaryltellurium (IV) dichlorides, R_2TeCl_2 with the Schiff base obtained by condensation of indole-2,3-dione with ethylenediamine.

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EXPERIMENTAL

Materials and methods

All the chemicals were of analytical reagent grade. All preparations were carried out under an atmosphere of dry N₂ as the compounds are sensitive to moisture. The solvents were dried by standard method before use. *p*-Methoxyphenyltellurium (IV) trichloride^{37,38}, bis(*p*-methoxyphenyl) tellurium (IV) dichloride^{38,39}, *p*-hydroxyphenyltellurium (IV) trichloride⁴⁰, bis(*p*-hydroxyphenyl)tellurium (IV) dichloride⁴⁰, 3-methyl-4-hydroxyphenyltellurium (IV) trichloride⁴¹ and bis(3-methyl-4-hydroxyphenyl)tellurium (IV) dichloride⁴¹ were prepared by the reactions of TeCl₄ with anisole/phenol/*o*-cresol as reported in the literature³⁷⁻⁴¹.

Preparation of ethylenediamine bis-(Isatin) Schiff base, [N, N'-Bis(indol-2-oxo-3-ylidene)-1,2-diaminoethane]

The Schiff base was prepared by the method reported by Khalifa and Hassaan¹⁵. A hot ethanolic solution of isatin (0.02 mol) was added to an ethanolic solution of 0.01 mol ethylenediamine under heating. The contents were refluxed for 2 hrs during, which time, a light brown product was precipitated. The mixture was cooled to room temperature and solid product was filtered off, washed with ethanol and recrystallized from DMF. Finally washed with diethyl ether and dried in vacuo over P₄O₁₀.

Preparation of complexes

Tellurium tetrachloride, aryltellurium (IV) trichlorides and diaryltellurium (IV) dichlorides, when reacted with ethylenediamine bis-(isatin) form 1:1 type of complexes as described below:

TeCl₄ (H₂L), RTeCl₃ (H₂L) and R₂TeCl₂ (H₂L)

These types of complexes were prepared by addition of hot methanolic solution of the EDBI (10 mmol in about 50 mL) to a solution of tellurium (IV) derivatives (10 mmol) in about 50 mL dry methanol with continuous stirring. The reaction mixture was refluxed on steam bath for 4 hrs. The excess solvent was distilled off to obtain the desired products, which were recrystallized from dry methanol and dried in vacuum desiccator over P₄O₁₀. The reactions were also repeated by addition of sodium methoxide until pH 7.3, but identical products were obtained unlike Sn(IV) and Zr(IV), where Schiff base acts as a binegative tetradentate ligand⁷.

Physical studies

Carbon, hydrogen and nitrogen analyses were obtained microanalytically from SAIF, Panjab University Chandigarh on a Thermo Finnigan CHNS analyser. Conductance measurements were carried out in DMSO at $25 \pm 2^\circ\text{C}$ with a dip type conductivity cell (cell constant = 1.017) on a microprocessor based conductivity bridge type MICROSIL.

Infrared spectra ($4000\text{-}400\text{ cm}^{-1}$) were recorded in KBr pellets at SAIF, Panjab University Chandigarh on a F. T. Infra-Red Spectrophotometer Model RZX (Perkin Elmer). Proton NMR Spectra were recorded in DMSO- d_6 using TMS as an internal reference on BRUKER AVANCE II 400 NMR spectrometer.

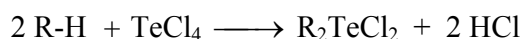
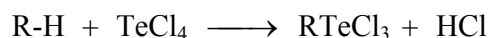
Antimicrobial studies

The compounds were prepared in 1000 and 500 ppm concentrations in acetone using Poison Plate Technique method⁴². Potato dextrose-agar (PDA) medium was prepared in flasks and sterilized. To this medium, a requisite quantity of solution was added and then the medium was poured into Petri plates in three replications. A culture of test fungus was grown on PDA for 6-7 days. Small disc (4 mm) of the fungus culture was cut with a sterile cork borer and transferred aseptically, upside-down in the center of Petri dishes containing the medium and fungicides. Plates were incubated at $25^\circ\text{C} \pm 1^\circ\text{C}$. Colony diameters were measured and data were statistically analysed.

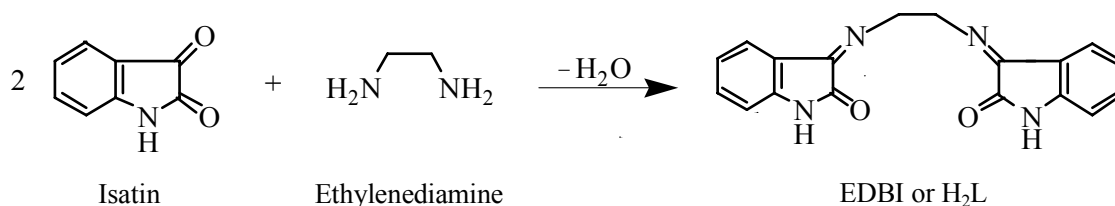
Antitubercular activity was evaluated in DMSO against *M. tuberculosis* H₃₇Rv using Microplate alamar blue assay (MABA) method^{43,44}. Antitubercular susceptibility test was performed in black, clear-bottomed, 96-well microplates (Packard Instrument Company, Meriden, Conn., USA) in order to minimize background fluorescence. Primary screening of the compounds for antitubercular activity has been conducted at $12.5\ \mu\text{g/mL}$.

RESULTS AND DISCUSSION

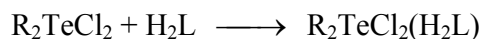
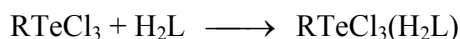
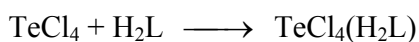
The formation of aryltellurium (IV) trichlorides and diaryltellurium(IV) dichlorides by the reactions of TeCl_4 with anisole³⁷⁻³⁹, phenol⁴⁰ and *o*-cresol⁴¹ (R-H) involves the electrophilic substitution of the aromatic ring by a chlorotellurium group at a position *para* to the methoxy/hydroxyl group.



Preparation of ethylenediamine bis-(isatin), (EDBI) Schiff base, by the reaction of isatin with ethylenediamine can be represented by following equation:



EDBI reacts with tellurium (IV) chloride, aryltellurium (IV) trichlorides and diaryltellurium (IV) dichlorides in 1:1 molar ratio to form the corresponding tellurium (IV) complexes:



All the tellurium (IV) complexes are colored solids, stable at room temperature and non-hygroscopic in nature. They are insoluble in non polar organic solvents, but are soluble in polar donor organic solvents like DMF, DMSO etc. The analytical data and physical properties of Schiff base and the tellurium (IV) complexes are presented in Table 1.

Conductance studies

The molar conductance, Λ_M at *ca.* 10^{-3} M data (Table 1) for the complexes (78.89-86.34 $\text{ohm}^{-1} \text{cm}^2 \text{mol}^{-1}$) indicate^{45,46} that the complexes are 2:1 electrolyte type in DMSO solution. This suggests that tellurium (IV) is present as cationic moiety, where as two chloride ions are as anions in the complexes. The Schiff base complexes of tellurium (IV), thus may be represented as $[\text{TeCl}_2(\text{H}_2\text{L})] \text{Cl}_2$, $[\text{RTeCl}(\text{H}_2\text{L})] \text{Cl}_2$ and $[\text{R}_2\text{Te}(\text{H}_2\text{L})] \text{Cl}_2$, respectively.

Infrared spectra

The important infrared spectral data of EDBI and its tellurium (IV) complexes are compiled in Table 2. The spectra of tellurium (IV) complexes are quite complex and thus, an attempt has been made to identify the donor sites by comparing the spectra of complexes with the parent ligand and tellurium (IV) chlorides.

Table 1: Analytical data, molar conductance and physical properties for ethylenediamine bis-(isatin) complexes of tellurium (IV)

Compd No	Complex (R)	Empirical formula (Formula wt.)	Colour (Yield %)	M. Pt. (°C) dec.	Analyses % Found (Calculated)				Δ_M at ca. $10^{-3} M$ $ohm^{-1} cm^2 mol^{-1}$ in DMSO	
					C	H	N	Te		
Schiff base	EDBI / H ₂ L	C ₁₈ H ₁₄ N ₄ O ₂ (318.22)	Light brown (88)	219-221	67.23 (67.94)	4.72 (4.40)	17.40 (17.61)	-	-	
I	TeCl ₄ (H ₂ L)	C ₁₈ H ₁₄ Cl ₄ N ₄ O ₂ Te (587.63)	Orange (79)	198-200	36.01 (36.79)	2.01 (2.38)	9.32 (9.53)	21.21 (21.71)	23.76 (24.13)	86.34
II	RTeCl ₃ (H ₂ L) (<i>p</i> -methoxyphenyl)	C ₂₅ H ₂₁ Cl ₃ N ₄ O ₃ Te (659.26)	Dark brown (81)	180-182	44.96 (45.55)	2.87 (3.19)	7.98 (8.50)	18.97 (19.36)	15.81 (16.13)	82.45
III	RTeCl ₃ (H ₂ L) (<i>p</i> -hydroxyphenyl)	C ₂₄ H ₁₉ Cl ₃ N ₄ O ₃ Te (645.25)	Light orange (84)	190-192	45.03 (44.67)	3.42 (2.95)	8.27 (8.68)	19.34 (19.78)	15.96 (16.48)	78.89
IV	RTeCl ₃ (H ₂ L) (3-methyl-4-hydroxyphenyl)	C ₂₅ H ₂₁ Cl ₃ N ₄ O ₃ Te (659.26)	Light brown (76)	138-140	44.89 (45.55)	2.97 (3.19)	8.01 (8.50)	19.09 (19.36)	15.79 (16.13)	83.71
V	R ₂ TeCl ₂ (H ₂ L) (<i>p</i> -methoxyphenyl)	C ₃₂ H ₂₈ Cl ₂ N ₄ O ₄ Te (730.88)	Light yellow (78)	144-146	52.98 (52.59)	3.35 (3.83)	7.31 (7.67)	17.80 (17.46)	9.21 (9.70)	79.23
VI	R ₂ TeCl ₂ (H ₂ L) (<i>p</i> -hydroxyphenyl)	C ₃₀ H ₂₄ Cl ₂ N ₄ O ₄ Te (702.86)	Brown (69)	110-112	50.87 (51.27)	3.01 (3.41)	7.44 (7.97)	17.88 (18.15)	9.65 (10.09)	84.56
VII	R ₂ TeCl ₂ (H ₂ L) (3-methyl-4-hydroxyphenyl)	C ₃₂ H ₂₈ Cl ₂ N ₄ O ₄ Te (730.88)	Dark brown (75)	132-134	52.12 (52.59)	3.42 (3.83)	7.34 (7.67)	17.03 (17.46)	9.16 (9.70)	81.65

Table 2: Important IR data (cm⁻¹) for the ligand (EDBI) and complexes

Compound No.	$\nu_{(N-H)}$	$\nu_{(C=O)}$	$\nu_{(C=N)}$
EDBI / H ₂ L	3269 sb	1740 s	1652 s
I	3192 s	1729 s	1615 s
II	3192 s	1728 s	1616 s
III	3193 s	1729 s	1614 s
IV	3194 mb	1727 s	1616 s
V	3195 m	1727 s	1616 s
VI	3190 b	1732 s	1619 s
VII	3192 mb	1728 s	1614 s

s = strong, m = medium, b = broad

The three strong bands appearing at 3269, 1740 and 1652 cm⁻¹ in the ligand may be assigned^{7,10,15} to stretching vibration modes ν_{N-H} , $\nu_{C=O}$ and $\nu_{C=N}$ of azomethine group, respectively.

In the IR spectra of TeCl₄(H₂L), RTeCl₃(H₂L) and R₂TeCl₂(H₂L) complexes, the bands of free ligand at 1740 cm⁻¹ and 1652 cm⁻¹ displayed shifts to lower wave numbers 1732-1727cm⁻¹ and 1619-1614 cm⁻¹, respectively indicating the involvement of oxygen atom of C=O group of isatin residue and nitrogen atom of azomethine group in complex formation. The ν_{N-H} band remains intact and appears at 3190-3195 cm⁻¹ in the complexes. The most important conclusion drawn from infrared spectral evidence is that the ethylenediamine-bis(isatin) is acting as chelating agent towards central tellurium atom as a neutral ONNO quadridentate ligand^{10,12}. The ν_{Te-O} and ν_{Te-N} could not be ascertained due to non-availability of Far IR data.

Proton magnetic resonance spectra

The ¹H NMR spectra of free ligand and its tellurium (IV) complexes were measured in DMSO-d₆ and the data are presented in Table 3.

The spectrum of free ligand can be resolved into three distinct regions: complex multiplet at 6.835-7.590 δ ppm corresponding to isatin skeleton, one singlet at 10.869 δ ppm corresponding to isatin NH residue and one singlet at 3.338 δ ppm due to ethylenic chain bridged protons.

Table 3: ^1H NMR spectral data of Schiff base (EDBI) and complexes

Compound No.	Chemical Shift, δ ppm in DMSO- d_6
EDBI / H_2L	3.338 (s, 4H, ethylenic chain bridged protons), 6.835-7.590 (cm, 8H, fused aromatic nuclei of isatin residue), 10.869 (s, 2H, NH)
I	3.409 (s, 4H, ethylenic protons), 6.904-7.584 (cm, 8H, aryl protons of isatin), 11.031 (s, 2H, NH)
II	3.473 (s, 4H, ethylenic protons), 3.84 (s, 3H, $-\text{OCH}_3$), 6.930-8.222 (cm, 12H, aryl protons of isatin and RTe), 11.105 (s, 2H, NH)
III	3.399 (s, 4H, ethylenic protons), 6.907-7.582 (cm, 12H, aryl protons of isatin and RTe), 8.355 (bs, 1H, phenolic OH), 11.071 (s, 2H, NH)
IV	2.186 (s, 3H, CH_3), 3.398 (s, 4H, ethylenic protons), 6.861-8.203 (cm, 11H, aryl protons of isatin and RTe), 9.392 (bs, 1H, phenolic OH), 11.104 (s, 2H, NH)
V	3.368 (s, 4H, ethylenic protons), 3.776 (s, 6H, OCH_3), 6.696-7.835 (cm, 16H, aryl protons of isatin and R_2Te), 11.03 (s, 2H, NH)
VI	3.464 (s, 4H, ethylenic protons), 6.769-7.836 (cm, 16H, aryl protons of isatin and R_2Te), 9.935 (bs, 2H, phenolic OH), 11.07 (s, 2H, NH)

s = singlet, cm = complex multiplet, bs = broad singlet. Spectra of compound number VII not well resolved due to poor solubility

The proton NMR spectra of complexes i.e. $\text{TeCl}_4(\text{H}_2\text{L})$, $\text{RTeCl}_3(\text{H}_2\text{L})$ and $\text{R}_2\text{TeCl}_2(\text{H}_2\text{L})$ display a downfield shift from 11.03 to 11.105 δ ppm, which is associated with the hydrogen of isatin NH residue. This behavior is related with a decrease of electron density and deshielding of NH proton, as a result of participation of the adjacent carbonyl group in coordination^{47,48}. Also, ethylenic protons resonating at 3.338 δ ppm in free ligand, appear at downfield side (3.368-3.473 ppm) in the complexes, indicating the flow of electron density from the azomethine nitrogens to the tellurium centre.

Independent assignments to the aryl protons of Schiff base and RTe/ R_2Te are not possible due to overlapping of signals in this region. Thus, proton magnetic resonance spectral studies support the foregoing IR spectral evidence of EDBI acting as a neutral quadridentate (ONNO) ligand in these complexes.

On the basis of conductance, IR and ^1H NMR spectral studies, a distorted octahedral environment around central tellurium atom may be suggested as shown in Fig. 1.

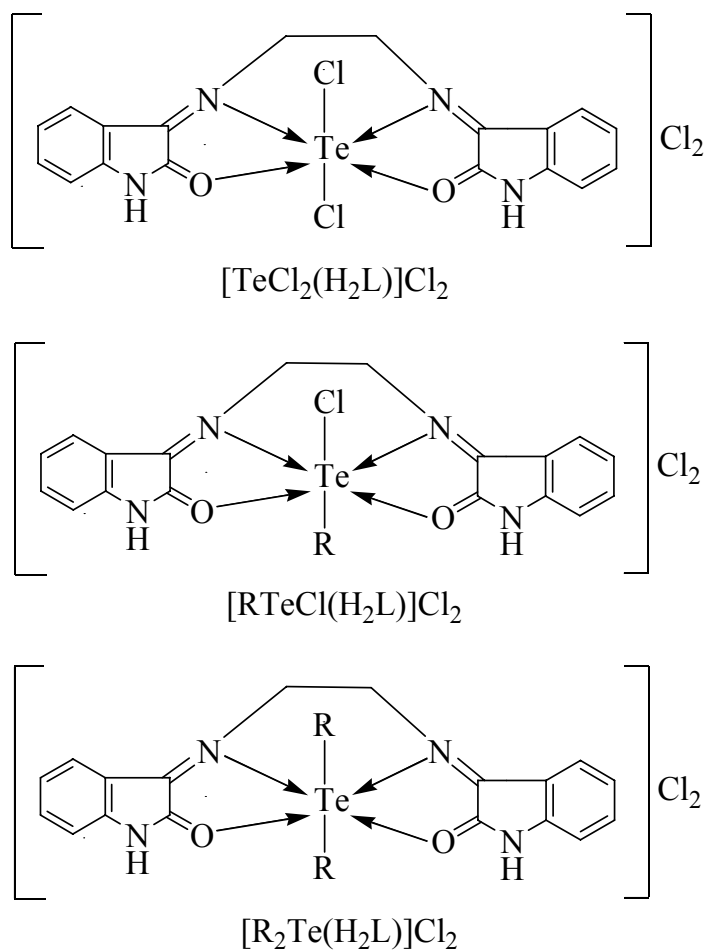


Fig. 1: Proposed structures of tellurium (IV) ethylenediamine bis-(isatin) Schiff base complexes

Biological studies

Ethylenediamine bis-(isatin) Schiff base (EDBI) and some of its complexes were evaluated for antifungal and antitubercular activity *in vitro*. Fungicidal activity data (Table 4) indicate that the compounds III and VI possess better antifungal activity against all the three pathogens, while other compounds show moderate to good activity towards these pathogens.

Table 4: Effect of concentration of Schiff base (EDBI) and complexes on the mean radial growth (cms) of fungus in vitro (Poison Plate Technique)⁴²

Compd. No.	<i>Rhizoctonia solani</i>		<i>Fusarium oxysporum</i>		<i>Colletorichum capsici</i>	
	1000 ppm	500 ppm	1000 ppm	500 ppm	1000 ppm	500 ppm
EDBI / H ₂ L	1.38	2.25	3.42	5.37	3.83	6.43
I	7.92	9.00	1.98	2.83	3.58	5.51
III	1.47	3.21	1.10	4.17	1.25	3.42
IV	6.15	8.64	1.40	2.67	2.67	3.25
V	7.02	9.00	3.58	5.58	3.00	4.95
VI	1.75	1.92	1.83	2.75	1.87	3.65
Check	9.00	9.00	8.67	8.67	7.67	7.67
CD%	0.78	1.21	0.91	0.92	1.06	1.28

CD% = Standard antifungal drug Fluconazol

Antitubercular activity data were compared with the standard drug Rifampin at 0.25 µg/mL concentrations, which showed 98% inhibition. The results are presented in Table 5, which indicate that the compounds III and VI were very much effective against *M. tuberculosis* at 12.5 µg/mL concentrations and showed 90-92% inhibition while the other compounds showed moderate to good activity against *Mycobacterium tuberculosis*.

Table 5: Antitubercular activity against *Mycobacterium tuberculosis* of Schiff base (EDBI) and complexes

Compound No.	Antitubercular activity	
	(H ₃₇ Rv) MIC (µg/mL)	% Inhibition
EDBI/H ₂ L	< 12.5	48
I	< 12.5	45
III	< 12.5	90
IV	< 12.5	85
V	< 12.5	83
VI	< 12.5	92
Rifampin	0.25	98

MIC-Minimum inhibition concentration in µg/mL

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