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Microwave Synthesis, Spectral Studies, Antimicrobial Approach, and Coordination Behavior of Antimony(III) and Bismuth(III) Compounds with Benzothiazoline[1]

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Abstract—The reaction of 2-hydroxy-N-phenylbenzamide with 2-aminobenzenethiol yielded 2-hydroxy-N-phenylbenzamidebenzothiazoline (H_2-Saly · BTZ/HO⌒N⌒SH). The reaction of H_2-Saly · BTZ with $PhSbCl_2$, $SbCl_3$, and $BiCl_3$ under varied reaction conditions (microwave, as well as conventional method) gave corresponding antimony(III) and bismuth(III) Schiff base compounds (substitution along with addition) in different coordination environments. These complexes were characterized by elemental analysis, IR and NMR (^1H and ^{13}C) spectral studies. The ligand was found to bifunctional tridentate, as well as monodentate for different starting materials of metal (Sb/Bi), as well as for different reaction conditions, hence, suitable coordination environments and pseudotrigonal bipyramidal geometry for the antimony and bismuth complexes have been proposed. Their biological activities have also been checked against many fungi and bacteria. The complexes were found to be more toxic than the corresponding ligand.

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INTRODUCTION

The present day industrialization has led to immense environmental deterioration. The increasing environmental consciousness throughout the world has put a pressing need to develop an alternate synthetic approach for biologically and synthetically important compounds. This requires a new approach, which will reduce the material and energy intensity of chemical processes and products, minimize or eliminate the dispersion of harmful chemicals in the environment in a way that enhances the industrially benign approach and meets the challenges of green chemistry [1]. Microwave-assisted synthesis is a branch of green chemistry. In other words, requirement of few amount of solvents coupled with high yields and short reaction times is often associated with the reactions of this type and makes these procedures very attractive for synthesis. In the present discussion, we describe the synthesis of the metal complexes with one Schiff base using microwave assisted technique. Benzothiazolines constitute an important class of $–NC_6H_4S–$ containing ligands. Usually benzothiazoline ring opening is catalyzed by the presence of a metal ion [2]. However, some reactions with benzothiazolines are reported [3] to form addition products also. In general biological activity of such type of ligands enhances considerably on complexation with the metal atom [4]. Benzothiazolines and their metal chelates have been found to exhibit fungicidal [5], bactericidal [5], antifertility [5], and pharmacological activities [6]. The coordination chemistry, biological effects, and toxicology of antimony and bismuth complexes, such as their requirements in pharmacological activities, are areas of increasing research interest [7, 8]. Antimony compounds exhibit a broad spectrum of biological activities [9], chemotherapeutic applications [10], and cytotoxic activities [11]. Bi(III) exhibits variable coordination numbers and often an irregular coordination geometry. In view of the diversified chelating behavior of benzothiazolines, as well as novel biological importance of antimony and bismuth complexes [12], it has been considered worthwhile to synthesize, characterize some new antimony(III) and bismuth(III) derivatives of benzothiazolines and to investigate their physicochemical and structural features as well as the biological activity. A comparative study has also been made on the basis of microwave radiation effects on compound synthesis, reports of which are rather few in the literature. In continuation of our work, we have studied the coordination behavior of benzothiazoline towards the Sb and Bi metals.

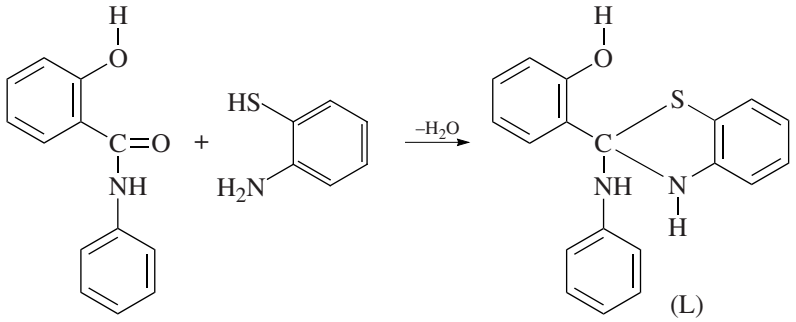
EXPERIMENTAL

The $PhSbCl_2$ was prepared according to the literature method [13]. 2-Hydroxybenzamide, 2-aminobenzenethiol, and trichlorobismuthane were purchased and used as such. All the chemicals were dried and

[1] The article is published in the original.

purified before use. All the preparations were done under anhydrous conditions. The purity was checked by thin layer chromatography.

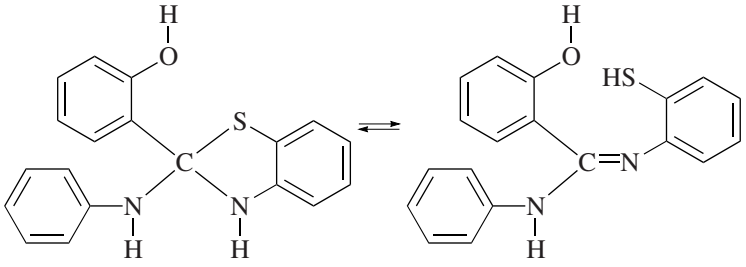
Synthesis of 2-hydroxy-N-phenylbenzamidebenzothiazoline (L) was carried out by the condensation of 2-hydroxy-N-phenylbenzamide with 2-aminothiophenol in a 1 : 1 molar ratio using ethanol.



The reaction mixture was stirred for 3–4 h, and the solid separated out was filtered, purified by recrystallization from ethanol, and dried *in vacuo*. Product is sandy brown, m.p. 110°C, FW (found/calcd) 298.3/320.4.

For $C_{19}H_{16}N_2SO$ (HO⌒N⌒SH)		
anal. calcd, %:	N, 8.7;	S, 10.0.
Found, %:	N, 8.1;	S, 9.8.

The parent ligand exists in the tautomeric forms (benzothiazoline ring as well as Schiff base):



(Ring form) (Azomethine form)

For the comparison purpose, two different routes were employed for the synthesis of the antimony and bismuth compounds (thermal and microwave) (Table 1).

Synthesis of the organo-, chloroantimony(III)-, and chlorobismuth(III)-complexes [PhSb(O⌒N⌒S) (I), SbCl(O⌒N⌒S) (II), and BiCl(O⌒N⌒S)] (III).

Microwave method [14]. For the synthesis of complexes, dichloromonophenylantimony(III) (or $SbCl_3$ or $BiCl_3$), and sodium salt of H_2-Saly · BTZ (prepared by adding the corresponding weight of sodium to 2-hydroxy-N-phenylbenzamidebenzothiazoline) in 5 ml of dry methanol in 1 : 1 molar ratio were irradiated inside a microwave oven at 700 W for about 5–8 min. The products were recovered from the microwave oven and dissolved in few ml of dry methanol. The white precipitate of sodium chloride formed during the course of the reaction was removed by filtration, and the filtrate was dried under reduced pressure. The resulting product was repeatedly washed with petroleum ether and then finally dried at 40–60°C/0.5 mmHg for 3–4 h. The purity was further checked by thinlayer chromatography using silica gelG. The details of these reactions and the analysis of the resulting products are recorded in Table 1.

Thermal method. These organo- and chloroantimony(III) and chlorobismuth(III) complexes were also synthesized by the thermal method. The reaction mixtures were heated under reflux for 10–15 h and filtered to remove NaCl and the solvent was removed by the same procedure mentioned above, which was adopted to get the complexes.

Synthesis of antimony trichloride and bismuth trichloride adducts MCl_3 · (HO⌒N⌒SH) (M = Sb(IV), Bi(V)).

Microwave method. In microwave-assisted synthesis, the reaction mixtures were taken in an open borosil beaker and then irradiated for 4–7 minutes. A drastic reduction in the reaction time was observed due to the rapid heating capability of microwaves. Finally, the adducts

Table 1. Comparison of data of microwave and conventional methods

Compound	Yield, %		Solvent, ml		Time	
	thermal	microwave	thermal	microwave	thermal, h	microwave, min
PhSb(O⌒N⌒S)	55	79	15	5	14	8
SbCl(O⌒N⌒S)	46	80	15	5	13	10
$SbCl_3 \cdot$ (HO⌒N⌒SH)	43	79	15	3	12	3
BiCl(O⌒N⌒S)	57	84	15	2	14	10
$BiCl_3 \cdot$ (HO⌒N⌒SH)	58	86	10	5	12	3

Table 2. Analytical and physical characteristics of the ligand and its antimony and bismuth derivatives

Compound	Reactant, g			Color	M.p., °C	Contents (found/calcd), %							M.w. (found/ calcd)
	starting material	ligand	Na			C	H	N	S	Cl	Sb	Bi	
HO⌒N⌒SH				Sandy brown	110			8.1/8.7	9.8/ 10.0				298.3/ 320.4
PhSb(O⌒N⌒S)	$PhSbCl_2$ (0.732)	0.869	0.125	Brown	178*	57.78/ 58.05	3.53/ 3.70	5.12/ 5.42	6.11/ 6.20		23.19/ 23.54		518.43/ 517.26
SbCl(O⌒N⌒S)	$SbCl_3$ (0.461)	0.648	0.093	Red	184	47.55/ 47.98	2.45/ 2.97	5.34/ 5.89	6.45/ 6.74	7.34/ 7.45	25.45/ 25.60		476.11/ 475.61
$SbCl_3 \cdot$ (HO⌒N⌒SH)	$SbCl_3$ (0.531)	0.746		Light yellow	169	42.26/ 42.85	2.97/ 3.03	5.23/ 5.26	5.88/ 6.02	19.43/ 19.97	23.91/ 23.86		533.12/ 532.53
BiCl(O⌒N⌒S)	$BiCl_3$ (0.823)	0.836	0.120	Gray	132	40.15/ 40.55	2.33/ 2.50	4.88/ 4.98	5.66/ 5.70	6.05/ 6.30		36.98/ 37.13	562.42/ 562.83
$BiCl_3 \cdot$ (HO⌒N⌒SH)	$BiCl_3$ (0.813)	0.826		Creamish	156	35.78/ 35.90	2.45/ 2.54	4.23/ 4.41	4.78/ 5.04	16.64/ 16.73		32.01/ 32.87	636.45/ 635.75

* Decomposition temperature.

were recovered from the microwave oven, washed repeatdly with *n*-hexane, then with petroleum ether, and finally dried at 40–60°C/0.5 mmHg for 3–4 h. The purity was further checked by TLC using silica gelG.

Thermal method. In this method, adducts were prepared under anhydrous conditions. Diethyl ether and carbon tetrachloride solution of $BiCl_3$ and $SbCl_3$, respectively, were added dropwise to a benzene solution of H_2-Saly · BTZ in a unimolar ratio at room temperature with constant stirring. A creamish to light yellow-colored precipitate was obtained after 12 h continuously stirring. The precipitate was filtered off and washed with the parent solvent twice or thrice and then with dry *n*-hexane. Finally, adducts were dried under reduced pressure to yield the desired product.

Analytical and physical characteristics of antimony and bismuth complexes are given in Table 2.

Physical measurements and analytical methods. The molecular weights were determined by the Rast camphor method. Sulfur and nitrogen were estimated gravimetrically (Messenger's method) as $BaSO_4$ and by Kjeldahl's method, respectively. Chlorine was determined by Volhard's method. Bismuth was estimated complexometrically [15]. Antimony was estimated by the oxidation of Sb(III) to Sb(V) on heating with $KMnO_4$, the excess of which was decolorized with H_2O_2. The remaining H_2O_2 was decomposed, and Sb(V) was then determined iodimetrically [16]. Electronic spectra of the complexes were recorded in methanol on a UV–160A Shimadzu spectrophotometer in the range 200–600 nm. Infrared spectra of the ligands and its complexes were scanned in the range 4000–200 cm^{-1} with the help of a model Nicolet Megna FTIR–550 spectrophotometer and a model FTIR–8400 S spectrophotometer on KBr pellets. The NMR spectra were recorded using a JEOL-AL–300 FT NMR spectrometer in DMSO-d_6 using TMS as the internal standard. The conductivity of the resulting derivatives was determined at room temperature in dry DMF by the Systronics conductivity bridge (Model 305) using a cell having a cell constant of 0.5 cm^{-1}. Carbon and hydrogen analyses were performed at the Central Drug Research Institute (Lucknow, India).

Antifungal screening. The antifungal activity was evaluated against *Fusarium oxysporum* and *Alternaria alternata* using the agar plate technique. The compounds were directly mixed with the medium in different concentrations. Controls were also run, and three replicates

Table 3. IR spectral data (cm^{-1}) of the ligand and its antimony(III) and bismuth(III) derivatives

Compound	ν(Ph–OH)	ν(>C=N)	ν(NH)	ν(M–N)*	ν(M–O)*	ν(M–S)*
HO⌒N⌒SH	3405		3310			
PhSb(O⌒N⌒S)		1620		400	515	380
SbCl(O⌒N⌒S)		1505		430	505	391
$SbCl_3 \cdot$ (HO⌒N⌒SH)	3310		3275	450		
BiCl(O⌒N⌒S)		1630		320	445	230
$BiCl_3 \cdot$ (HO⌒N⌒SH)	3404		3260	332		

* M = Sb or Bi.

were used in each case. The linear growth of the fungus was recorded by measuring the diameter of the fungus colony after four days. The amount of growth inhibition in each of the replicates was calculated by the equation: $100 \times (C–T)\ C^{-1}$, where C and T are the diameters of the fungus colony in the control and the test plates, respectively [17].

Antibacterial screening. Antibacterial activity was evaluated against *Pseudomonas aeruginosa* and *Escherichia coli* by the paper disc plate method. The nutrient agar medium (0.5% peptone, 0.15 yeast, 0.15 beef extract, 0.35 NaCl, and 0.13% KH_2PO_4) in distilled water (1000 cm^3) was autoclaved for 20 min at 15 *psi* before inoculation. The 5-mm diameter paper discs of Whatman no. 1 were soaked under different solutions (500 and 1000 ppm) of the compounds, dried, and then placed in the Petri plates previously seeded with the test organisms. The plates were incubated for 24 h at 28 ± 2°C, and the inhibition zone around each disc was measured [17].

RESULTS AND DISSCUSSION

The elemental analysis and spectral data are consistent with the formulation of compounds I–V. The unimolar reactions of $SbCl_3$, $PhSbCl_2$, and $BiCl_3$ with the sodium salt of the H_2-Saly · BTZ in a methanol solution proceed with the formation of M←N, M–S, and M–O bonds, yielding the substitution products. The reaction proceeds as shown in Eqs. (1)–(2):

$$PhSbCl_2 + HO⌒N⌒SH + 2Na \xrightarrow{MeOH} \longrightarrow PhSb(O⌒N⌒S) + 2NaCl, \quad (1)$$
(**I**)

$$MCl_3 + HO⌒N⌒SH + 2Na \xrightarrow{MeOH} \longrightarrow MCl(O⌒N⌒S) + 2NaCl. \quad (2)$$
(M = Sb(II), Bi(III))

In another type of reactions, metal chlorides ($SbCl_3$ and or $BiCl_3$) react with the benzothiazoline in 1 : 1 molar ratio. The reaction proceeded with the formation of molecular adducts. It is shown below in Eqs. (3):

$$MCl_3 + HO⌒N⌒SH \xrightarrow{CCl_4/\text{diethyl ether}} MCl_3 \cdot (HO⌒N⌒SH), \quad (3)$$

where M = Sb(IV), Bi(V), and HO⌒N⌒SH is the donor set of the ligand.

The resulting colored solids are monomeric in nature, as evidenced by their molecular weight determinations, and are soluble in most of common organic solvents. Their low molar conductivity values (8–10 $Ohm^{-1}\ cm^2\ mol^{-1}$) show that they are nonelectrolyte in nature. The unimolar reaction of H_2-Saly · BTZ with $SbCl_3$ and $BiCl_3$ in CCl_4/diethyl ether solvents at room temperature resulted in the precipitation of simple addition compounds. Liberation of HCl gas was not observed even under reflux. These observations show that the benzothiazoline ring is not opened during the reaction. These addition compounds are creamish-colored to yellow solid powders. They are soluble in common organic solvents and on heating tend to decompose with charring.

The UV spectrum of the benzothiazoline HO⌒N⌒SH consists of two broad bands centered at 260 and 310 nm characteristics of the cyclic forms of the ligand. These bands arise out of π–π* (benzenoid) transitions. The band in the range of 320–398 nm due to the *n*–π* electronic transition of the azomethine group is observed in the spectra of the substitution complexes **I**–**III**, which remains absent in the free ligand, thus proving the ring form of the benzothiazoline present in L.

The IR spectral data of the synthesized compounds are represented in the Table 3. Absence of the ν(SH) mode at 2610–2540 cm^{-1} and the presence of ν(NH) mode at 3310 cm^{-1} in the spectrum of the ligand HO⌒N⌒SH indicates the presence of the benzothiazoline ring structure [18] in the ligand. The IR-spectra of the complexes **I**–**III** show the disappearance of the NH stretching band. In these complexes, the phenolic OH stretching band also disappeared which was present at ~3405 cm^{-1} in the spectrum of the ligand, indicating the M–O bond formation during complexation. Some new bands observed in the regions 1630–1505, 400–450, 505–515, 380–391, 320–332, 230–265, and 445 cm^{-1} for

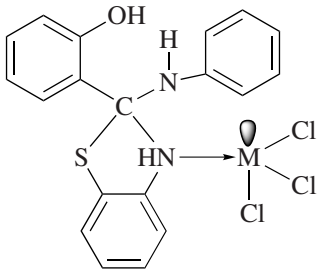
Table 4. ^{1}H NMR spectral data (δ, ppm) of the ligand and its antimony(III) and bismuth(III) derivatives*

Compound	Ph–OH (bs.)	–NH (free) (bs.)	–NH (ring) (bs.)	Aromatic (m.)
HO⌒N⌒SH	12.0	10.60	5.20	6.54–7.59
PhSb(O⌒N⌒S)		10.58		6.58–7.65
SbCl(O⌒N⌒S)		10.60		7.26–8.03
$SbCl_3$ · (HO⌒N⌒SH)	12.0	10.61	6.25 (deshielded)	6.58–8.06
BiCl(O⌒N⌒S)		10.57		6.71–8.05
$BiCl_3$ · (HO⌒N⌒SH)	12.1	10.62	6.17 (deshielded)	6.81–7.95

* bs. – broad singlet; m. – multiplet.

ν(>C=N), ν(Sb←—N) [19], ν(Sb–O) [20], ν(Sb–S) [21], ν(Bi←—N) [22], ν(Bi–S) [23], and ν(Bi–O) [24], respectively. The band at 450–470 cm^{-1} may be assigned to ν(Sb–Ph) [14] vibrations in PhSb(O⌒N⌒S) complex. This may be interpreted in terms of the benzothiazoline ring opening and the formation of Schiff base complexes by the rearrangement of the ring.

In the case of the adducts IV and V, the presence of ν(NH) mode of vibration indicates that the benzothiazoline ring remains intact [25] in the said adducts. This band due to ν(NH) mode appears with the lowering of ~20–35 cm^{-1} in its position in the synthesized adducts. This indicates the participation of the NH group in bonding. This is further supported by the appearance of a new M←—N band. This confirms the coordination in these adducts, taking place through nitrogen only and also showing the monodentate behavior of the H_2-Saly · BTZ ligand. It is shown below:



1 : 1 Adduct,
where M = Sb or Bi atom

The ^{1}H NMR spectral data are represented in the Table 4. The signal observed at δ 5.20 ppm due to the –NH proton in the spectrum of HO⌒N⌒SH is found to be absent in the complexes **I–III** indicating the deprotonation of the NH proton during complex formation. The phenolic (Ar–OH) proton appears at δ 12.0 ppm in the benzothiazoline and disappears in its complexes, showing the deprotonation of phenolic proton and confirming the M–O bond formation. This clearly indicates the tridentate behavior of the H_2-Saly · BTZ (L). In the spectra of compounds **IV** and **V**, NH proton becomes deshielded and shows a downfield shift. This supports the formation of the M←—NH bond during adduct formation. No appreciable shift has been observed in the position of the phenolic proton (Ar–OH) signal on complexation indicating that this group does not participate in bonding.

The ligand (HO⌒N⌒SH) shows multiplets in a region of δ 6.54–8.06 ppm attributable to aromatic protons, which appear almost in the same position in their respective complexes.

A comparative study of the ^{13}C NMR spectra of the benzothiazoline ligand with the antimony and bismuth(III) derivatives provides useful information about the mode of bonding. In the spectra of complexes **I–III**, a downfield shift of the >C–N carbon signal, which appears at ~162.54 ppm, confirming the formation of M←—N=C (Sb/Bi) by the rearrangement of benzothiazoline ring. A downfield shift in the position of the Ph–C–O group on complexation indicates the M–O bond formation (M = Sb/Bi). The substituted phenyl ring carbons are observed in the range δ 125.13–177.36 ppm. The –NC_6H_4S– carbon signals are observed in the range δ 127.16–150.56 ppm. A new set of signals was observed in ranges of δ 142.36, 124.72, 127.96, 128.98, 127.34, and 123.26 ppm for complex I, assigned to the phenyl ring carbons attached to antimony atom.

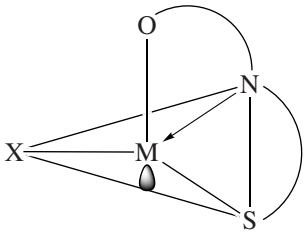
In the ^{13}C NMR spectra of the addition compounds IV and V, a small shift was observed in the position of the >C–N signal as compared to the free ligand. This indicates the involvement of nitrogen atom of the benzothiazoline in bonding. No appreciable shift has been observed in the position of >C–OH group carbon on complexation indicating that this group does not participate in bonding.

In view of the monomeric nature of these derivatives and bifunctional tridentate **I–III** as well as monodentate (adducts **IV** and **V**), behavior of the ligand in the compounds as evident from the observed spectral data (Tables 3 and 4), the following structure in which the central atom (Bi/Sb) aquires pseudotrigonal bipyramidal

Table 5. Antifungal screening and antibacterial screening data of the ligand and its antimony(III) and bismuth(III) derivatives

Compound	Average antifungal screening (%) (inhibition zone (mm) after 4 day; concentration in ppm)						Average antibacterial screening (%) (inhibition zone (mm) after 24 h; concentration in ppm)			
	Fusarium oxysporum			*Alternaria alternata*			*Escherichia coli*		*Pseudomonas aeruginosa*	
	50	100	200	50	100	200	500	1000	500	1000
HO⌒N⌒SH	34	47	69	37	48	68	7	9	6	9
PhSb(O⌒N⌒S)	42	54	85	43	52	81	9	11	9	13
SbCl(O⌒N⌒S)	41	51	80	42	51	71	8	10	7	11
$SbCl_3 \cdot$ (HO⌒N⌒SH)		49	71	35	53	64	7		9	
BiCl(O⌒N⌒S)	48	59	87	49	61	82	10	14	10	15
$BiCl_3 \cdot$ (HO⌒N⌒SH)	42	50	70	47		74	8	10		12

geometry appears to be highly plausible. It is shown below:



1 : 1 complex,

where M = Sb or Bi atom, X = Ph/Cl and HO⌒N⌒SH/H_2-Saly · BTZ ligand.

The ligand L and its metal complexes have been screened *in vitro* for their antibacterial and antifungal activities. The results are indicative of the fact that these compounds exhibit antimicrobial properties. These data are represented in the Table 5. Antimicrobials can attack various targets in microorganism, as a consequence of which the organisms are either destroyed or have their growth inhibited. Since the complexes inhibit the growth of microorganisms, it is assumed that the production of the enzymes is being affected and, hence, the microorganisms are unable to utilize the food for themselves, or the intake of ion decreases and, consequently, the growth ceases. At lower concentrations when the enzyme leaches out, the growth of the microorganism is arrested, although a very small amount of enzyme is being produced, but the amount is sufficient to suffice the need of the microorganism to grow, while higher concentrations destroy the enzyme mechanisms by blocking any of the metabolism pathways (*viz.*, lipid, carbohydrate, and aminoacids), and the organism dies [26]. Furthermore, enzymes seem to be a natural target for inorganic drugs, since metals play a key structural role for many enzymes, such as zinc metalloenzymes perturbing an endogenous metal that is vital to enzymatic action can render such an enzyme inactive. These disturbances can arise from actions such as coordination of exogenous ligands to the metal, substitution of the metal, or removal of the metal. Inorganic complexes can also affect nonmetalloenzymes. Metals can coordinate to active site residues to block substrate interaction or coordinate to residues outside the active site to affect structural integrity. The coordination ability of metal also holds the attractive promise of forming stronger attachments through covalent and ionic bonds [27].

The results reveal that there is a considerable increase in the toxicity of the complexes as compared to the ligands. On giving a closer look at these results, a common feature, which appears is that the bioactivity enhances due to the following points.

1. The chelation reduces the polarity and increase the lipophilic nature of the central metal atom, which subsequently favors its permeation through the lipid layer of the cell membrane. This can be well ascribed to Tweedy's chelation theory [28].

2. Solubility and concentration of the compounds also play an important role in biological activity. It is seen that lower concentration of compounds can check the sporulation in fungi, and a higher concentration inhibits the growth of organisms almost completely.

3. The enhanced activity of the complexes depends on fineness of the particle size of the metal ion and the presence of the bulkier organic moieties. Furthermore, halogen atoms directly attached to metal ion increases toxicity to some extent and, when this halogen atom is being replaced by a bulky ligand moiety, it enhances the activity of the whole molecule to a considerable extent [29].

4. The toxicity of antibacterial compounds against different species of bacteria depends either on the difference in ribosomes, or the impermeability of the cell to the antimicrobial agent [30].

CONCLUSIONS

Microwave irradiation is an efficient and environmentally-benign method to accomplish various inorganic syntheses to afford products in higher yields in shorter reaction periods.

N-Phenylbenzamidebenzothiazoline (HO⌒N⌒SH) ligand behaves as tridentate, as well as monodentate,

with different starting materials of the metal under different reaction conditions.

Antimony(III) and bismuth(III) complexes obtained by 1 : 1 molar reaction with sodium salt of the ligand were found to be tetracoordinate but, due to stereochemically active lone pair of electrons present on the metal atom, pseudotrigonal bipyramidal geometry have been tentatively proposed for the said complexes.

Antimicrobial activity of the complexes and the ligand L showed that the former are more active than the parent ligand.

The data given in Table 5 reveal that the PhSb(O⌒N⌒S) (I) and BiCl(O⌒N⌒S) (III) complexes were found to be more toxic than the other complexes and the bismuth complexes display better results than the antimony complexes.

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REFERENCES

1. Kidwai, M. *Pure Appl. Chem.*, 2001, vol. 73, p. 147.
2. Agrawal, M., Tandon, J.P., and Mehrotra, R.C., *Inorg. Nucl. Chem.*, 1981, vol. 43, p. 1070.
3. Chauhan, H.P.S., Srivastava, G., and Mehrotra, R.C., *Indian J. Chem.*, 1984, vol. 23, p. 436.
4. Fahmi, N. and Singh, R.V., *Transition Met. Chem.*, 1994, vol. 19, p. 453.
5. Kanoongo, N., Singh, R.V., Tandon, J.P., and Goyal, R.B., *J. Inorg. Biochem.*, 1990, vol. 38, p. 57.
6. Pandey, T. and Singh, R.V. *Main, Group Met. Chem.*, 2000, vol. 23, p. 346.
7. Klapotke, T., *Biol. Met.*, 1988, vol. 1, p. 69.
8. Smith, K.A., Deacon, G.B., Jackson, W.R., et al., *Metal Based Drugs*, 1998, vol. 5, p. 295.
9. *Comprehensive Coordination Chemistry*, Wilkinson, G., Ed., Oxford (UK): Pergamon, 1987.
10. Haiduc, I. and Silvestru, C., *Organometallics in Cancer Therapy*, Boca Raton, FL: CRC, 1989, vol. 3.
11. Khosa, M.K., Mazhar, M., Ali, S., et al., *Turk. J. Chem.*, 2006, vol. 30, p. 345.
12. *Handbook on Metals in Clinical and Analytical Chemistry*, Iffland, R., Seiler, H.G., Sigel, A., and Sigel, H., Eds., New York: Marcel Dekker, 1994.
13. Nunn, M., Sowerby, D.B., and Wesolek, D.M., *J. Organometal. Chem.*, 1983, vol. 45, p. 251.
14. Mahajan, K., Fahmi, N., and Singh, R.V., *Indian J. Chem., A*, 2007, vol. 46, p. 1221.
15. Vogel, A.I., *A Text Book of Quantitative Inorganic Analysis*, London: Longmans, 1989.
16. *Comprehensive Analytical Chemistry, B*, Wilson, C.L. and Wilson, D.W., Eds., New York: Elsevier, 1960, vol. 1.
17. Garg, R., Fahmi, N., and Singh, R.V., *Russ. J. Coord. Chem.*, 2008, vol. 34, p. 203.
18. Singh, R.V., Mittal, S.P., Swami, M., and Mahajan, K., *Int. J. Chem. Sci.*, 2007, vol. 5, p. 1417.
19. Chremos, G.N. and Zingaro, R.A., *J. Organomet. Chem.*, 1970, vol. 2, p. 647.
20. Bril1, T.B. and Campbel1, N.C., *Inorg. Chem.*, 1973, vol. l2, p. 1884.
21. Sharma, R.K., Singh, Y.P., and Rai, A.K., *Synth. React. Inorg. Met.-Org. Chem.*, 2001, vol. 31, p. 405.
22. Agoes, L., Burford, N., Cameron, T.S., *e*t al., *J. Am. Chem. Soc.*, 1996, vol. 118, p. 3225.
23. Briand, G.G., Burford, N., Cameron, T.S., and Kwaitkoswki, W., *J. Am. Chem. Soc.*, 1998, vol. 120, p. 177.
24. Sharma, M.K., Sharma, M., Singh, A., and Mehrotra, R.C., *Indian J. Chem., A*, 2001, vol. 40, p. 1226.
25. Chauhan, H.P.S., Shrivastava, G., and Mehrotra, R.C., *Indian J. Chem., A*, 1984, vol. 23, p. 434.
26. Gaur, S., Maanju, S., Fahmi, N., and Singh, R.V., *Main Group Met. Chem.*, 2005, vol. 28, p. 293.
27. Louie, A.Y. and Meade, T.J., *Chem. Rev.*, 1999, vol. 99, p. 2711.
28. Tweedy, B.G., *Phytopathology*, 1964, vol. 55, p. 910.
29. Garg, R., Saini, M.K., Fahmi, N., and Singh, R.V., *Transition Met. Chem.*, 2006, vol. 31, p. 362.
30. Lawrence, P.G., Harold, P.L., and Francis, O.G., *Antibiotic and Chemotherapy*, 1980, vol. 5, p. 1597.