

Journal of Applicable Chemistry

2016, 5 (5): 1127-1136 (International Peer Reviewed Journal)



Synthesis, Spectroscopic Characterization and Biologically Investigations of Some New Coordination Compounds of Arsenic (III) with Biologically Active Carbohydrazones using Green Solvent

Sunita Choudhary, Sarita Varshney and A. K. Varshney*

*Department of Chemistry, University of Rajasthan, Jaipur-302004, INDIA

Email: anilakv123@rediffmail.com

Accepted on 23rd August 2016

ABSTRACT

Some new coordination compounds of arsenic (III) having general formula $[AsCl_3(L)]$ (where L = carbo hydrazone ligand) have been synthesized by the interaction of arsenic trichloride with carbohydrazone ligands with the ratio of 1:1 (metal-ligand) using dry methyltetrahydrofuran as a reaction medium. The newly synthesized complexes were further characterized by elemental analysis, molecular weight determinations and conductivity measurement. Plausible structures are proposed on the basis of spectral studies viz., IR, UV-Vis and NMR. The biological activities of carbohydrazones and their newly synthesized complexes have been screened in vitro against some bacterial and fungal strain to assess their growth inhibitory potency. Most of the metal complexes exhibit more antibacterial and antifungal activities than the free carbohydrazone ligands against these organisms.

Keywords: Carbohydrazones, spectral analysis, biological activities.

INTRODUCTION

Carbohydrazones are known to be a class of versatile ligands in coordination chemistry due to their ease of synthesis and diversity as well as structural possibilities. According to literature, they have been also as potent antimicrobial, antioxidant, therapeutic, anticonvulsant, cytotoxic and pharmacological as well as catalytic agents [1-4]. Carbohdrazone ligands synthesize by using carbohydrazide having enormous biological applications due to their oxygen and nitrogen donor atoms. Carbohydrazones exist in equilibrium of various tautomers due to unique structural features which greatly affect their chelating ability [5].

The coordination chemistry of metal complexes using green solvents is very interesting because of their structural diversity and multiple advantages in diverse fields such as low toxicity, low miscibility, easily biodegradable under environmental conditions, high boiling point and easy to recycle after use [6]. During the course of present investigation, we have synthesized five ligands and their corresponding metal derivatives using green solvent *viz*. methyltetrahydrofuran.

The chemistry of arsenic (III) compounds is important due to their wide range of applications. Interest in the chemistry of these compounds is growing continuously because of structural variations. Arsenic compounds are also known to possess significant biological applications. Some of arsenic (III) complexes with Schiff base showed effective antimicrobial activities [7-10]. In addition to these applications, complexes of arsenic are also commercially important in heterogeneous catalysis [11]

Continuing earlier research [14] on biologically active complexes, systematic studies on the binding of carbohydrazones to As(III) metal ion lead to the conception to develop new and efficient complexes, play a vital role in a vast number of biological process. In view of these facts, here we were thus motivated to undertake a systematic study of preparation and structural characterization of some newly synthesized complexes formed with carbohydrazones (LH) and As (III) ion. The carbohydrazones used in this study are shown in fig. 1. In addition, the biological studies were applied to the free ligands and their corresponding As (III) complexes against different bacterial, fungal strains using inhibitory zone diameter.

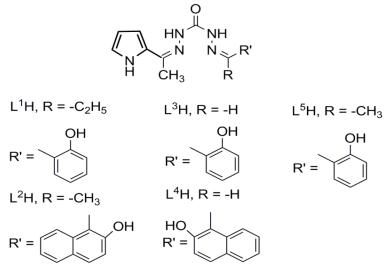


Fig. 1. Carbohydrazones used as ligands in this work.

MATERIALS AND METHODS

Materials: Solvents were dried by standard methods before use. $AsCl_3$ (Aldrich) has been used as supplied. The ligands used in this study have been synthesized in the laboratory using standard method reported in the literature [4].

Physical measurements: Melting points were recorded on Gallenkamp melting point apparatus. IR spectra have been recorded using 8400 Shimadzu FT-IR Spectrophotometer using KBr pellets in the range of 4000-400 cm⁻¹. ¹H and ¹³C NMR in CDCl₃ solution were recorded with a JEOL-FT A1 300 MHz spectrometer using TMS an internal reference. Micro analytic data for (C, H, N) were recorded on a Coleman 5612 analyzer. Chlorine was estimated by Volhard's method [12]. Arsenic was estimated iodimetrically [13]. The UV-Vis spectra of the carbohydrazones and their As (III) complexes were recorded on a 1800 Shimadzu UV spectrophotometer in the range of 200-800 nm. Molecular weights were determined by Rast method. The conductivity of the resulting Coordination compounds 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} . All the compounds have been synthesized using similar method therefore the synthetic and analytical data of the prepared complexes have been summarized in table 1.

Compound	M.P. (°C)	Color	% Elemen Found (Ca	tal analysis alcd.)	M:L in dry methyltetra hydrofuran	Mol. Wt Found (Calcd.)		
			С	Н	Ν	As		
As[(C ₁₆ H ₁₈ N ₅ O ₂)(Cl) ₃]	135	Yellow	39.11 (38.93)	3.69 (3.67)	14.73 (14.19)	15.25 (15.18)	1:1	491.32 (493.62)
As[(C ₁₉ H ₁₈ N ₅ O ₂)(Cl) ₃]	141	Orange	43.36 (43.09)	3.44 (3.42)	13.31 (13.22)	14.23 (14.15)	1:1	526.35 (529.65)
As[(C ₁₄ H ₁₄ N ₅ O ₂)(Cl) ₃]	150	Radish	36.31 (36.12)	3.04 (3.03)	15.12 (15.04)	16.18 (16.09)	1:1	463.13 (465.57)
As[(C ₁₈ H ₁₆ N ₅ O ₂)(Cl) ₃]	147	Dark Yellow	42.19 (41.93)	3.14 (3.12)	13.67 (13.58)	14.62 (14.53)	1:1	512.37 (515.62)
$As[(C_{15}H_{16}N_5O_2)(Cl)_3]$	130	Yellow	37.83 (37.57)	3.38 (3.36)	14.70 (14.60)	15.73 (15.62)	1:1	476.27 (479.59)

 Table 1. Physical data of As(III) complexes of carbohydrazones

Synthesis of carbohydrazones: Carbohydrazones have been synthesized by refluxing the reaction mixture of hot ethanolic solution of carbohydrazide and hot ethanolic solution of suitable carbonyls *viz.*, salicylaldehyde, 2-hydroxy-1-nephthaldehyde, *o*-hydroxyacteophenone, 2-hydroxy-1-acteonaphthone, *o*-hydroxypropiophenone and 2-acetylpyrrole in 1:1:1 molar ratio for 2 h. The products obtained after the evaporation of the solvent were filtered and recrystalized from ethanol.

Synthesis of As (III) complexes with carbohydrazones: The complexes of As(III) have been synthesized by the reactions of arsenic trichloride with carbohydrazones ligands in 1:1 molar ratios in dry methyltetrahydrofuran. The reaction mixture was heated under reflux on a fractioning column for 8-10 h. After the completion of reaction, the excess solvent was removed under reduced pressure to yield coloured viscous liquid. The newly synthesized products were repeatedly washed with *n*-hexane.

Antimicrobial activity: Antimicrobial activity of chemically derived compounds was studied. Three bacterial and fungal strains were selected for the primary screening.

Microorganisms Used: Clinical laboratory bacterial isolates of *Bacillus subtilis*, *Staphylococcus aureus*, and *Escherichia coli* and fungal isolates *viz.*, *Fusarium oxysporium*, *Trichoderma reesei* and *Penicillium funiculosum* were collected from the stock cultures of Microbiology Laboratory, SMS Medical College Jaipur, India.

Preparation of samples: The 10 mg mL⁻¹ of samples was dissolved in DMSO and further dilutions were made for calculating *MIC* value.

Culture and maintenance of bacteria: Pure cultures obtained from SMS Medical College Jaipur, India were used as indicator organisms. These bacteria were grown in Nutrient agar medium (prepared by autoclaving 8 % Nutrient agar of Difco-Laboratories, Detroit, USA, in distilled water at 15 lbs psi for 30 min) by incubating at 37 °C for 48 h. Each bacterial culture was further maintained on the same medium after every 48 h of transferring. A fresh suspension of test organism in saline solution was prepared from a freshly grown agar slant before antimicrobial assay.

Determination of Antibacterial Assay: In vitro antibacterial activity of the samples was studied against gram positive and gram negative bacterial strains by the agar well diffusion method [14]. Streptomycin was used as reference antibacterial agent (control). Mueller Hinton agar no. 2 (Hi Media, India) was used as the bacteriological medium. The Mueller Hinton agar was melted and cooled to 48-50 °C and a

standardized inoculums (1.5×108 CFU mL⁻¹, 0.5 McFarland) was then added aseptically to the molten agar and poured into sterile Petri dishes to obtain a solid medium. Wells were prepared in the seeded agar plates. The test compound (100 µL) was introduced in the well (6 mm). The plates were incubated overnight at 37 °C. The antimicrobial spectrum of the chemical compounds was determined for the bacterial species in terms of zone sizes around each well. The diameters of zone of inhibition produced by the agent were compared with those produced by the commercial control antibiotics, streptomycin. For each bacterial strain controls were maintained where pure solvents were used instead of the chemical compound. The control zones were subtracted from the test zones and the resulting zone diameter was measured with antibiotic zone reader to nearest mm. The experiment was performed three times to minimize the error and the mean values are presented.

Determination of Antifungal Assay: Anti fungal activity of the experimental plant was investigated by agar well diffusion method [15]. Ketokenazole was used as reference antifungal agent. The yeasts and saprophytic fungi were sub cultured on to Sabouraud's dextrose agar, SDA (Merck, Germany) and respectively incubated at 37 °C for 24 h and 25 °C for 2-5 days. Suspensions of fungal spores were prepared in sterile PBS and adjusted to a concentration of 106cells m L⁻¹. The plates were dried at room temperature for 15 min. Wells of 10 mm in diameter and about 7 mm apart were punctured in the culture media using sterile glass tube. 0.1 ml of several dilutions of fresh extracts was administered to fullness for each well. Plates were incubated at 37 °C. After incubation of 24 h bioactivities were determined by measuring the diameter of inhibition zone (in mm). All experiments were made in triplicate and means were calculated.

RESULTS AND DISCUSSION

The condensation reaction of carbohydrazide with suitable carbonyls in 1:1:1 molar ratio yields carbohydrazones. The ligands used in this study have been reported [4]. As(III) complexes of carbohydrazones have been synthesized by the reaction of arsenic trichloride with carbohydrazone ligands in 1:1 molar ratios, in refluxing dry methyltetrahydrofuran gave corresponding coordination complexes of As(III) (Scheme 1). After the completion of reactions, the excess solvent was removed under reduced pressure and then complexes were filter off to yield coloured solid product. Suitable crystals were not obtained under experimental conditions.



Scheme 1. Synthetic route for the preparation of As (III) complexes.

$\mathbf{R} = -\mathbf{C}_2\mathbf{H}_5$	complex – 1
$\mathbf{R} = -\mathbf{H}$	complex – 3
$R = -CH_3$	complex – 5
$R = -CH_3$	complex - 2
R = -H	complex – 4

After removing the solvent under reduced pressure, colored viscous compounds were obtained. The purity of these complexes was checked by TLC using silica gel-G as adsorbent in DMF as the solvent. Monomeric nature of these coordination complexes of As(III) demonstrated by molecular weight measurements. The molar conductance of the complexes (10^{-3} M) in dry DMF lies in the $11-14 \text{ ohm}^{-1} \text{ cm}^2 \text{ mol}^{-1}$ range, indicating that they are nonelectrolyte in nature due to no counter ion in the proposed structure of the complexes. The analytical data of carbohydrazones and their metal complexes are given at table 1, in a satisfactory agreement with the calculated values.

UV-Vis spectra: Most detailed information about the electronic structure of a compound, obtain by UV-Vis spectra. The UV-Vis spectra of the free ligands and their complexes were recorded using ethanol at room temperature. The UV spectrum of carbohydrazones (LH) showed two intense bands at 225-280 nm and 345-415 nm which belong to $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ transition respectively (Table 2). The band at 225-280 nm attributed to $\pi \rightarrow \pi^*$ transitions appear almost at the same positions in the spectra of their arsenic complexes. The band observed at 345-415 nm for free ligands shows a hypsochromic shift (310-375 nm) in the spectra of the arsenic due to coordination of azomethine nitrogen to the metal atom [16].

Cmpd.	UV-Vis (nm)	bands	Complexes	UV-Vis.bands (nm)
$L^{1}H$	225, 380		$As[(C_{16}H_{18}N_5O_2)(Cl)_3]$	220, 310
$L^{2}H$	235, 385		$As[(C_{19}H_{18}N_5O_2)(Cl)_3]$	232, 315
L ³ H	265, 345		$As[(C_{14}H_{14}N_5O_2)(Cl)_3]$	263, 335
L^4H	255, 400		$As[(C_{18}H_{16}N_5O_2)(Cl)_3]$	254, 340
L ⁵ H	280,415		$As[(C_{15}H_{16}N_5O_2)(Cl)_3]$	273, 350

 Table 2 UV-Vis Spectra of (HL) and their metal complexes

Infrared Spectroscopic Study: The IR spectra of complexes and their corresponding ligands were recorded as KBr pellets. A comparative study of the IR spectra of complexes and their corresponding ligands give useful information regarding bonding patterns of ligands. The reaction of carbohydrazide with suitable carbonyls produced carbohydrazones (LH). This reaction was followed by appearance of a characteristic new band in the range 1605-1625 cm⁻¹, which is due to frequency of the free azomethine groups, $v_{C=N}$ are utilized to confirm the structures of (HL) [17]. This band splits into two sharp bands at 1585-1610 cm⁻¹ and 1580-1600 cm⁻¹ on complexation of azomethine nitrogen to metal ion whereas the other one is due to uncoordinated nitrogen of azomethine group. The bands due to $v_{C=N}$ were shifted to lower frequencies [18] which reveals the bonding nature of the corresponding ligands and structural chemistry. A broad peak due to v_{OH} of –OH group appear in the region 3415-3450 cm⁻¹ in the spectra of ligands.

In spectra of metal complexes this band is shifted to lower frequencies indicates the involvement of oxygen of this group in bonding with metal. In the spectra of arsenic complexes new absorption bands of medium to weak intensity in regions 415–430 and 465–480 cm⁻¹ due to $v_{As \leftarrow N}$ and $v_{As - O}$ vibrations, respectively [19]. Some additional medium to strong intensity bands have also been observed in the region 1585–1595 cm⁻¹ and 985-995 cm⁻¹ in the spectra of arsenic complexes for v_{C-O} and v_{N-N} shows a small shift towards lower wave number as compared their position as a result of complex formation, showing chelation of oxygen atom to the As atom. In the IR spectra of these complexes the v_{As-CI} frequencies could not be observed, as the spectra have been recorded in the range of 4000–400 cm⁻¹. IR spectral data are systemized at table 3.

					mpronos:			
Compd.	$\nu_{C=N}$	v_{Bi-O}	ν_{Bi-N}	v_{As-O}	v_{As-N}	v_{N-N}	v_{C-O}	ν _{OH}
L ¹ H	1605	-	-	-	-	985	1585	3425-3450
$L^{2}H$	1615	-	-	-	-	987	1587	3420-3440
L ³ H	1620	-	-	-	-	991	1591	3415-3435
$L^{4}H$	1625	-	-	-	-	993	1593	3417-3430
L ⁵ H	1610	-	-	-	-	995	1595	3422-3445
$As[(C_{16}H_{18}N_5O_2)(Cl)_3]$	1598	-	-	465	415	997	1609	3325-3350
$As[(C_{19}H_{18}N_5O_2)(Cl)_3]$	1605	-	-	475	430	1007	1615	3330-3360

Table 3 Characteristic stretching vibration frequencies (cm⁻¹) located at FT-IR oh (LH) and their metal complexes.

As[(C ₁₄ H ₁₄ N ₅ O ₂)(Cl) ₃]	1600	-	-	470	420	1003	1605	3335-3355
As[(C ₁₈ H ₁₆ N ₅ O ₂)(Cl) ₃]	1597	-	-	480	425	1015	1612	3315-3335
As[(C ₁₅ H ₁₆ N ₅ O ₂)(Cl) ₃]	1600	-	-	468	418	1005	1607	3330-3345

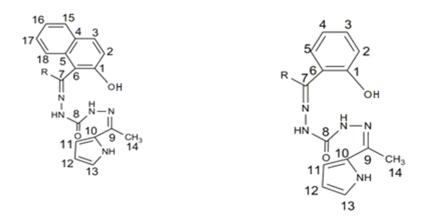
¹**H NMR:** The ¹H NMR data of ligands and their As(III) complexes have been recorded in CDCl₃ (Table 4). The –NH proton of the ligands, attached to >C=O group resonates at $\delta \sim 11.25$ ppm (S) and the signal in the region $\delta \sim 7.50$ -8.30 ppm was assigned to –NH proton of the pyrole ring present in the free ligands and their metal complexes. These signals remain unaltered in the spectra of the complexes, suggesting its non-involvement in complexation. In the ¹H NMR spectra of ligands, the signal due to the -OH proton of the ligands appears at $\delta \sim 11.20$ -12.55 ppm (S), was shifted downfield in the spectra of the corresponding As(III) complexes clearly suggests chelation of the ligand moiety through the phenolic group with the arsenic atoms in the complexes ($\delta \sim 11.75$ -13.10 ppm). In the case of the ligands, the proton signal for the methyl protons [-C(CH₃)=N] and azomethine protons [-CH=N] in the region $\delta \sim 1.75$ -1.85 ppm (S) and $\delta \sim 8.25$ -8.45 ppm, shifts downfield in the spectra of corresponding arsenic complexes on account of its deshielding, which is attributed to the donation of the lone pair of electrons by the azomethine nitrogen to the metal atom. The ligands show a complex multiplet in the region $\delta \sim 6.10$ -7.55 ppm for the aromatic protons which remains at almost the same position in the same spectra of the metal complexes.

Table 4 H NMR data for t	the ligand	s and for	their corresp	onding metal complexes
Comp. No.	-HCN	-OH	-CH ₃ CN	Aromatic protons
L ¹ H	8.30	11.20	1.75	6.15-7.20
$L^{2}H$	8.45	12.25	1.80	6.10-7.15
L ³ H	8.35	11.50	1.85	6.65-7.34
$L^{4}H$	8.30	12.55	1.83	6.10-7.55
L ⁵ H	8.25	11.55	1.79	6.78-7.35
As[(C ₁₆ H ₁₈ N ₅ O ₂)(Cl) ₃]	8.75	12.35	1.89	7.05-7.60
As[(C ₁₉ H ₁₈ N ₅ O ₂)(Cl) ₃]	9.42	12.95	1.90	7.15-7.50
As[(C ₁₄ H ₁₄ N ₅ O ₂)(Cl) ₃]	9.39	12.45	2.00	7.60-7.95
As[(C ₁₈ H ₁₆ N ₅ O ₂)(Cl) ₃]	9.24	13.05	1.97	7.35-7.80
$As[(C_{15}H_{16}N_5O_2)(Cl)_3]$	9.45	12.10	1.99	7.45-7.85

¹³C NMR Spectra: ¹³C NMR spectra of the arsenic (III) derivatives suggested the bonding through nitrogen and oxygen atom of ligand towards central metal atom. The ¹³C NMR spectral data for carbohydrazones and its corresponding As(III) complexes are reported in table 5. The shifting in the position of resonance of carbon attached to -OH group suggests the bonding of phenolic oxygen to the As(III) ion. The signal due to the carbon atom attached to the azomethine group in the ligands appears at $\delta \sim 170.8-173.1$ ppm. The shifting of azomethine (>C=N) carbon signal in the spectra of complexes ($\delta \sim 163.5-165.10$ ppm) indicate that the azomethine nitrogen has been involved in coordination with the As(III) ion, further confirms the complexation. The signal for the carbonyl carbon (C=O) present in the arsenic complexes appears in the range of $\delta \sim 160.30-165.20$ ppm.

 Table 5 ¹³C NMR data for the ligands and for their corresponding metal complexes

							(Chemical	shift valu	ιe in δ pp	m							
LH	C-1	C-2	C-3	C-4	C-5	C-6	C-7	C-8	C-9	C-10	C-11	C-12	C-13	C-14	C-15	C-16	C-17	C-18
$L^{2}H$	154.6	115.4	128.9	118.8	128.1	119.2	173.7	168.8	175.3	143.1	11.5	12.3	12.9	121.4	125.3	119.7	126.4	119.4
L^4H	153.5	114.3	126.5	116.7	124.4	116.5	170.8	166.4	172.7	140.1	10.4	10.5	12.0	122.7	123.6	120.4	125.9	120.7
L^1H	154.3	115.2	128.5	118.1	127.5	118.3	173.4	168.6	174.8	142.4	11.4	11.9	12.5	12.1	-	-	-	-
L ³ H	153.1	113.8	125.4	116.3	124.6	115.9	171.3	165.9	172.4	139.5	10.1	10.3	12.2	11.5	-	-	-	-
L⁵H	156.3	117.5	130.7	120.3	129.5	119.7	175.1	170.3	176.6	144.3	12.1	12.7	13.4	12.9	-	-	-	-



APPLICATIONS

In vitro antimicrobial assay: The *in vitro* antibacterial activity of the ligands and their complexes were screened against three bacterial strains by well diffusion method using streptomycin as reference they showed significant potent activity. The susceptibility of bacterial strain toward the compounds was estimated by measuring the size of inhibition zone diameter. It is evident from the results that the antibacterial activity of some of the metal complexes is higher than the free ligands and lesser than the standard against all the bacteria tested. In case of antibacterial activity carbohydrazones and their metal complexes were found to be active. Antifungal activity of the ligands and their complexes were screened against three fungal strains by well diffusion method using ketokenazole as reference they showed some potent activity. The antimicrobial results are systemized at table 6. It is, however, known that the chelating tends to make carbohydrazones act as more powerful and potent bacteriostatic agents, thus, inhibiting the growth of bacteria and fungi more than the parent carbohydrazones.

Cmpd.	Bacterial strain 20 ml 40 ml 60 ml 80 ml Fu B. subtilis 8 10 14 16 F. S. aureus 10 13 14 17 T. E. coli 9 14 14 18 P. B. subtilis 10 12 14 17 F.	Antifungal activit	Antifungal activity zone of inhibition (mm)							
		20 ml	40 ml	60 ml	80 ml	Fungal strain	20 ml	40 ml	60 ml	80 ml
	B. subtilis	8	10	14	16	F. oxysporium	nill	nill	10	13
$L^{1}H$	S. aureus	10	13	14	17	T. reesei	nill	10	15	18
	E. coli	9	14	14	18	P. funiculosum	8	10	14	16
	B. subtilis	10	12	14	17	F. oxysporium	9	10	13	14
$L^{2}H$	S. aureus	11	13	16	18	T. reesei	8	11	14	15
	E. coli	9	12	15	16	P. funiculosum	10	12	15	18
	B. subtilis	8	11	13	15	F. oxysporium	nill	10	14	17
L ³ H	S. aureus	10	12	14	17	T. reesei	8	9	11	13
	E. coli	11	12	14	16	P. funiculosum	9	12	14	16
	B. subtilis	9	10	12	14	F. oxysporium	8	11	11	13
L^4H	S. aureus	9	11	13	15	T. reesei	8	10	11	15
	E. coli	10	12	14	17	P. funiculosum	9	10	12	14
L ⁵ H	B. subtilis	9	10	13	15	F. oxysporium	9	10	13	16
	S. aureus	10	12	14	16	T. reesei	8	9	12	15
										1133

Table 6. Antimicrobial results of carbohydrazones and their metal complexes

	E. coli	11	13	14	15	P. funiculosum	Nill	8	12	16
	B. subtilis	10	12	13	14	F. oxysporium	8	10	12	16
As(L)Cl ₃	S. aureus	11	13	14	16	T. reesei	9	11	13	15
	E. coli	9	10	13	15	P. funiculosum	10	12	14	118
	B. subtilis	10	12	14	16	F. oxysporium	9	12	15	18
As(L)Cl ₃	S. aureus	11	14	16	17	T. reesei	10	12	16	18
	E. coli	12	13	18	19	P. funiculosum	12	15	18	16
	B. subtilis	10	13	15	19	F. oxysporium	10	13	16	18
As(L)Cl ₃	S. aureus	12	14	16	18	T. reesei	11	14	16	19
	E. coli	13	15	18	16	P. funiculosum	12	13	18	16
	B. subtilis	11	13	15	19	F. oxysporium	11	13	16	19
As(L)Cl ₃	S. aureus	12	14	15	17	T. reesei	12	14	15	17
	E. coli	12	13	17	16	P. funiculosum	10	13	18	16
	B. subtilis	11	13	15	19	F. oxysporium	11	13	15	19
As(L)Cl ₃	S. aureus	10	14	15	17	T. reesei	10	14	16	17
	E. coli	12	13	18	16	P. funiculosum	12	13	18	16
Strept	tomycin (for	bacteria)	– Inhibi	ition zone -	- 20mm;	Ketokenazole (for	fungi) -	- Inhibitio	n zone –	22mm.

Streptomycin (for bacteria) – Inhibition zone – 20mm; Ketokenazole (for fungi) – Inhibition zone – 22mm. *Standered

Structure of metal complexes: Spectroscopic investigations of these complexes suggested bidentate nature of ligand moiety towards arsenic ion. Molecular weight measurements show the monomeric nature of these complexes. Carbohydrazone ligand binds to arsenic through a nitrogen and oxygen atom of phenolic group. (Figure 2) In all metal complexes, in view of the presence of one bidentate chelate rings and three chlorine atoms, a pentacoordinated environment around the metal ion may be tentatively proposed for these complexes.

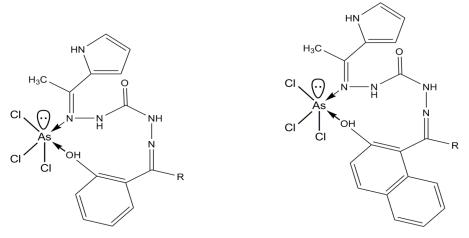


Fig. 2 Plausible structures of the metal complexes

CONCLUSIONS

During the course of present investigation, we have synthesized some coordination compounds of arsenic (III) with carbohydrazone ligands and determined their structure and physical properties. Bonding mode of ligands was suggested by IR and NMR (¹H and ¹³C) spectral studies. Ligands act as bidentate chelating agent in these complexes, which coordinate with As (III) ions through azomethine nitrogen and phenolic oxygen. A distorted octahedral geometry of these complexes of the type [As $Cl_3(L)$] (L = carbohydrazone ligand) was suggested by elemental analysis, spectral investigations *viz.*, IR, UV-Vis and NMR (¹H and ¹³C). These compounds show non electrolytic nature. Furthermore, these complexes were found to have significant antibacterial and antifungal activity. Since the carbohydrazones revealed to be poorly bioactive,

their arsenic (III) complexes might act by increasing the metals' bioavailability due to the lipophilic character of the ligands.

ACKNOWLEDGEMENTS

Authors are thankful to Head, Department of Chemistry, University of Rajasthan, Jaipur for providing necessary laboratory facilities. Sunita Choudhary is thankful to UGC, New Delhi, India for financial assistance as junior research fellow.

REFERENCES

- [1] A. Shrivastav, N. K. Singh, P. Tripathi, T. George, J. R. Dimmock, R. K. Sharma, Copper(II) and manganese(III) complexes of N'[(2hydroxy phenyl) carbonothioyl] pyridine2carbohydrazide: novel therapeutic agents for cancer, *Biochim.*, **2006**, 88, 1209-1216.
- [2] Z. H. Cohen, H. Pervaz, K. M. Khan, C. T. Supuran, Organometallic based antibacterial and antifungal compounds: transition metal complexes of 1, 1'diacetylferrocenederived thiocarbohydrazone, carbohydrazone, thiosemicarbazone and semicarbazone, *J. Enzyme Inhib. Med. Chem.*, **2005**, 20, 81-88.
- [3] R. Gupta, Synthesis and Spectral Studies of Organoarsenic (III) Complexes of Schiff Bases, J. *Applicable Chem.*, **2015**, 4 (1), 300-307.
- [4] S. Choudhary, S. Varshney, A. K. Varshney, Synthesis and Spectroscopic Characterization of Some New Coordination Compounds of Organotin (IV) with Carbohydrazones J. Applied Chem. Res, 2016, 10, 99-110.
- [5] G. M. Abu El-Reash, O. A. El Gammal, A. H. Radwan, Molecular structure and biological studies on Cr(III), Mn(II) and Fe(III) complexes of heterocyclic carbohydrazone ligand *Spectrochim. Acta Part A*, **2014**, 121, 259-267.
- [6] D. Clarke, M. A. Ali, A. A. Clifford, Reactions in unusual media, *Curr. Top. Med. Chem.*, **2004**, 4, 729-771.
- [7] P. Sreedhar, G. Srinivas, R. M. Raju Synthesis, Characterization and Antibacterial Activity of Novel Schiff Bases Bearing Thiazole Ring, *J. Applicable Chem*, **2016**, 5 (4), 776-782.
- [8] A.L.V. Kumar Reddy, Synthesis and Antibacterial Activity of Some Novel Hydrazone Derivatives of Anacardic acid Linked with 1, 4-Disubstituted 1,2,3-Triazole Ring. J. Applicable Chem., 2016, 5 (4), 783-791.
- [9] L. Dawara, S. C. Joshi, R. V. Singh, Synthesis, Characterization, and Antimicrobial and Antispermatogenic Activity of Bismuth(III) and Arsenic(III) Derivatives of Biologically Potent Nitrogen and Sulfur Donor Ligands, *Int. J. Inorg. Chem.*, **2012**, 2012, 1-9.
- [10] W. H. Jr. Miller, H. M. Schipper, J. S. Lee, J. Singer, S. Waxman, Mechanism of action of arsenic trioxide, *Cancer Res.*, 2002, 62, 3893-3903.
- [11] V. Le Boisselier, E. Dunach, E. Postel, Bismuth(III) Catalyzed Oxidative Cleavage of Aryl Epoxides Substituent Effects on the Kinetics of the Oxidation Reaction, J. Organomet. Chem., 1994, 482, 119-123.
- [12] H. S. Yadav, P. Choudhary, S. Varshney, A. K. Varshney, Electrochemical and Biochemical Investigations of Some New Coordination Compounds of Ni (II) with Azomethines Derived from Sulfa Drugs J. Applicable Chem., 2016,5 (2), 466-475.
- [13] A. I. Vogel, A Text Book of Quantitative Inorganic Analysis, 5th ed. (Longmans, London, 1989).
- [14] A. I. Vogel, A Textbook of Quantitative Chemical Analysis, Pearson Education Ltd.: Thames Polytechnique, London, UK, 6th edition, 2006.
- [15] C. Perez, M. Paul, P. Bazerque, Antibiotic assay by agar well diffusion method, *Biol. Med. Exp.*, **1990**, 15, 113-115.
- [16] S. Bonjar, S. Aghighi, N. A. Karimi, Antibacterial and anti fungal survey in plants used in indigenous herbalmedicine of South East Regions of Iran, *J. Biol. Sci.*, **2005**, 4 (4) 405-412.

- [17] A. Phor, synthesis and spectroscopic studies of biologicallyactive organoantimony(III) complexes of nitrogen donor ligands, *Phosphorus, Sulfur, and Silicon*, **1995**, 25(10), 1685-1697.
- [18] C. Jayabala Krishnan, K. Natarajan, Synthesis, Characterization and Biological Activities of Ruthenium(II) Carbonyl Complexes Containing Bi-Functional Tridentate Schiff Bases, Synth. React. Org. Met.-Org. Chem., 2001, 31, 983.
- [19] G. N. Chremos, R. A. Zingaro, Correlation of metal-chalcogen stretching frequencies of some trialkyl group Va chalcogenides, *J. Organomet. Chem.*, **1970**, 22, 647-651.

AUTHORS' ADDRESSES

1. Sunita Choudhary

2.

Department of Chemistry, University of Rajasthan, Jaipur, India. Email: sunitachoudhary555@gmail.com, Mob no: 9782857276

Dr Sarita Varsheny Associate Professor, Department of Chemistry, University of Rajasthan, Jaipur, India.

Email: saritavarshney@rediffmail.com, Mob no: 09414455822

3. Dr A.K. Varsheny

Associate Professor, Department of Chemistry, University of Rajasthan, Jaipur, India. Email: anilakv123@rediffmail.com, Mob no: 09414922215