Synthesis, characterization and antimicrobial activities of hydroxytriazenes and their Co(II) complexes

Prabhat K. Baroliya*, Mangilal Regar, R.S. Chauhan and A.K. Goswami
Coordination Chemistry Lab, Department of Chemistry, Mohanlal Sukhadia University, Udaipur, India

Síntesis, caracterización y actividades antimicrobianas de los hidroxitriacenos y sus complejos de Co(II)

Síntesi, caracterització i activitats antimicrobianes dels hidroxitriacenos i els seus complexos de Co(II)

Recibido: 1 de enero de 2014; aceptado: 25 de febrero de 2014

RESUMEN

El presente artículo describe la síntesis de los hidroxitriacenos sustituidos por flúor y cloro y de sus complejos de Co(II). Además estos compuestos junto con sus complejos de Co(II) han sido caracterizados debidamente por los análisis de CHN, IR, ^{1}H NMR y espectro de masas. La composición de los complejos se ha determinado mediante la proporción molar y el método de Job. Se ha sugerido una geometría tetraédrica para todos los complejos. Los compuestos se han utilizado para los estudios de actividad biológica frente a 4 cepas de bacterias y 6 cepas de hongos. Los valores MIC (concentración inhibitoria mínima) variaban entre 25 µg/mL y 50 µg/mL.

Palabras clave: Hidroxitriaceno, actividad antimicrobiana, Cobalto(II).

SUMMARY

The present paper describes synthesis of flouro chloro substituted hydroxytriazenes and their Co(II) complexes. Further these compounds along with their Co(II) complexes have been duly characterized by CHN, IR, ^{1}H NMR and Mass spectral analysis. Complex composition has been determined by Mole ratio and Job's method. A tetrahedral geometry has been suggested for all the complexes. The compounds have been used for biological activity studies against 4 bacteria and 6 fungal strains. The MIC (Minimum inhibitory concentration) values varied between 25 µg/mL to 50 µg/mL.

Key words: Hydroxytriazene; antimicrobial activity, Cobalt(II).

RESUM

El present article descriu la síntesi dels *hidroxitriacenos* substituïts per fluor i clor i dels seus complexos de Co(II). A més aquests compostos juntament amb els seus complexos de Co(II) han estat caracteritzats degudament per les anàlisis de CHN, IR, ^{1}H NMR i espectre de masses. La composició dels complexos s'ha determinat mitjançant la proporció molar i el mètode de Job. S'ha suggerit una geometria tetraèdrica per a tots els complexos. Els compostos s'han utilitzat per als estudis d'activitat biològica enfront de 4 ceps de bacteris i 6 ceps de fongs. Els valors MIC (concentració inhibitòria mínima) variaven entre 25 µg/ml i 50 µg/ml.

Mots clau: *Hidroxitriaceno*, activitat antimicrobiana, Cobalt(II).

*Correponding author:
Email: pbaroliya@yahoo.com

1. INTRODUCTION

The chemistry of hydroxytriazenes having –N(OH)-N=N– group and its derivatives has been studied for more than last 50 years due to their complexing and potential pharmacological properties. Hydroxytriazenes play important role in bioinorganic chemistry, as they form stable complex with most of the transition metal ions.[1-6] They present a variety of bioactivities including antibacterial[7-9], antifungal[8,9], anti-inflammatory[10,11], analgesic[12], wound healing[13] and insecticidal activities.[14] Further, hydroxytriazenes have shown good results in photo induced green synthesis of azo dyes.[15] Medicinally important metal complexes have become an interesting research field since the discovery of cis-platin.[16,17] Cobalt complexes are known to possess a broad spectrum of biological activities. In view of the versatile importance of hydroxytriazenes and cobalt complexes, we here in describe synthesis, characterization and antimicrobial activity of hydroxytriazenes and their Co(II)complexes. To determine the stoichiometry of the Co(II)–hydroxytriazene complexes in solution a spectrophotometric study has been undertaken using acetone as solvent. The composition of complexes was determined using mole ratio method and Job's method.

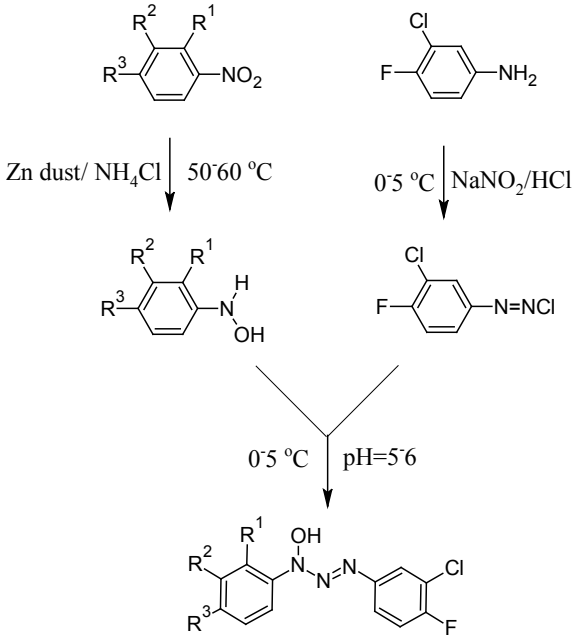
2. EXPERIMENTAL

2.1 Materials and methods

All chemicals used were of Analytical Reagent grade and employed directly without further purification. All melting points were determined on an Electrothermal Gallenkamp apparatus and are uncorrected. Perkin-Elmer C, H, N analyzer (model-2400) was used. The IR spectra were obtained on KBr pellets using a Perkin Elmer Spectrum RX1 (4000-450 cm^{-1}) stretching and bending bends are presented in terms of ν and δ, respectively. ^1H NMR spectra were recorded on Bruker AVANCE III 500 MHz NMR spectrometer in DMSO-D_6 with TMS as an internal standard. The chemical shifts are reported in parts per million (ppm) expressed in δ units and coupling constant (J) values are given in Hertz (Hz). The mass spectrum was obtained on Perkin-Elmer Sciex Triple Quadrupole LC/MS/MS Mass Spectrometer (Model-016932) using Ion Spray source. The samples were dissolved in DMSO-d_6. The reactions were monitored by thin layer chromatography (TLC), carried out on silica gel glass plates.

2.2 Synthesis of hydroxytriazenes

Four hydroxytriazenes were synthesized using standard method reported in literature.[18] In this method three steps are involved: (1) Nitro compounds were reduced with Zn dust in the neutral medium (NH_4Cl) in water-alcohol mixture at 50-60 °**C** to obtain respective hydroxylamine. The resulting mixture was used in coupling process. (2) In this step 3-chloro-4-fluoro aniline was diazotized with sodium nitrite at 0-5 °**C** in acidic medium under constant stirring. (3) Hydroxyl amine product from step-1 was coupled with aryl diazonium salt product from step-2, in acetate buffer medium of 5-6 pH at 0-5 °**C** temperature. A crude product obtained was purified and recrystallized with acetone. Scheme for synthesis of all hydroxytriazenes have been described in Figure.1. Purity of each hydroxytriazene was checked by TLC, melting point detection and characterized by CHN, IR, NMR and MASS spectral analysis.



	-R^1	-R^2	-R^3
1. BPHT	-H	-H	-H
2. o-TPHT	-CH_3	-H	-H
3. m-TPHT	-H	-CH_3	-H
4. p-TPHT	-H	-H	-CH_3

Figure 1. *Synthetic route and molecular structure of all synthesized hydroxytriazenes*

2.3 Synthesis of complexes

Co(BPHT), Co(o-TPHT), Co(m-TPHT) and Co(p-TPHT) were synthesized according to a reported procedure with slight modification.[23] The requisite metal (1.5 mmol) suspended in 5 mL of water and was added to the solution of hydroxytriazene, (3 mmol in acetone). The mixture was stirred for 1 h at 40 °**C** temperature on a water bath. The coloured precipitate so formed was filtered, washed with water and cold acetone and dried in oven. All the cobalt complexes were characterized by elemental analyses, IR, conductance and magnetic susceptibility measurements and, thermogravmeteric analysis.

2.4 Electronic spectra and molar conductivity measurements

The electronic absorption spectra were recorded by using a Shimadzu UV-1700 using DMSO as solvent. Conductivity of cobalt complexes was measured in freshly prepared acetone solutions and obtained using a Digital conductivity bridge (model: DI-909) and a dip type cell calibrated with KCl solution.

2.5 Magnetic susceptibilities measurement

The Magnetic susceptibilities of complexes were determined on Gouy balance model 7550 at 23 °**C**. The diamagnetic corrections were made by Pascal's constant and $Hg[Co(SCN)_4]$ was used as a calibrant.

2.6 Thermogravmetric analysis

The thermogravimetric analysis (TGA) provides authentic information regarding the presence and absence of water molecules in the coordination sphere of the complex. TGA

was carried out on a Perkin–Elmer model TGS-2 instrument.

2.7 Procedure for Study of Complex Formation in Solution

In order to determine the stiochiometric Co(II):HT ratio of the complex in acetone, the molar ratio and Job's continuous variation methods were employed.[24] Complex formation of TPHT with Co(II) has not been studied due to poor solubility. In the continuous variations method Co(II) and hydroxytriazenes solutions, 10^{-3} M, each were prepared and mixed. For the mole ratio method the [Co(II)] was maintained to 3×10^{-5} M while the hydroxytriazenes concentration were varied from 1 to 30×10^{-5} M. In both methods, spectra were recorded on a 400–800 nm range spectrophotometer.

2.8 Antibacterial activity

The synthesized hydroxytriazenes and their metal complexes were screened against *E. coli* (ATCC9637), *Pseudomonas aeruginosa, Staphylococcus aureus, K. Pneumoniae* where Norfloxacin and Ciprofloxacin were used as a standard drug. The bacterial strains were grown on nutrient agar at 37 °**C**. After 24 h of incubation, bacterial cells were suspended in normal saline containing Tween 20 at 0.05% at a concentration of approximately $1.0–2.0\times10^{7}$ cells/mL by matching with 0.5 McFarland standards. The activity of hydroxytriazenes and their cobalt complexes was determined as per NCCLS[25,26] protocol using Mueller Hinton broth (Becton Dickinson, USA) in 96-well tissue culture plates. Proper growth control, drug control and the negative control were adjusted onto the plate. Compounds were dissolved in DMSO at a concentration of 1 mg/mL and 20 mL of this was added to each well of 96-well tissue culture plate having 180 mL Mueller Hinton broth. From here the solution was serially diluted resulting in twofold dilution of the test compounds in subsequent wells. 100 mL of McFarland matched bacterial suspension was diluted in 10 ml of media and then 100 mL of it was added in each well and kept for incubation. The maximum concentration of compounds tested was 50 mg/mL. The micro-titer plates were incubated at 35 °**C** in a moist, dark chamber and MICs were recorded spectrophotometrically after 24 h using SOFT max Pro 4.3 Software (Molecular Devices, Sunnyvale, USA).

2.9 Antifungal activity

The antifungal activity of hydroxytriazenes and their metal complexes were evaluated against *Candida albicans, Cryptococcus neoformans, Sporothrix schenckii, Trichophyton mentagrophytes, Aspergillus fumigatus and Candida parapsilosis* (ATCC-22019). The activity of compounds was determined as per NCCLS[25,26] protocol in 96-well tissue culture plates. Proper growth control, drug control and the negative control were adjusted onto the plate. The fungal strains are grown in Sabouraud Dextrose Agar slants at 35 °**C**. The activity of compounds is determined by twofold microbroth dilution method using RPMI 1640 (1.04 g per 100 mL) buffered with (3-N-morpholino) propanesulphonic acid (MOPS; 3.47 g per 100mL RPMI) in 96-well tissue culture plates. The antifungal activity of the Bivittoside-D was determined by twofold microbroth dilution method as per guide lines of NCCLS. Briefly, the compounds were dissolved in dimethyl sulphoxide (DMSO, 10%) to get a stock (10 mg/mL) solution. The MIC of each compound was determined against test isolates by using broth micro dilution technique as described by the NCCLS. MICs of standard antifungal compound (Ketoconazole) and the compounds were measured in 96% well tissue culture plate using RPMI 1640 media buffered with MOPS (Sigma Chemical Co.). Starting incubation at 35 °**C** in a moist, dark chamber, MIC values were recorded spectrophotometrically after 24 hours.

3. RESULTS AND DISCUSSION

3.1 Characterization of the hydroxytriazenes:

The hydroxytriazene ligands (Scheme 1) were prepared by standard method as reported in literature.[18] Elemental analyses indicated that the ligand has the molecular formula as given below. The IR spectrum of the hydroxytriazenes shows a weak broad band at 3440 cm^{-1}, assigned to ν^{O-H} and a band at 3200 assigned to ν^{N-H}. The spectrum shows some stretching vibrational bands at 1425 ± 10, 1324 ± 10, 1234 ± 10, and 1,202 ± 5 cm^{-1}, attributed to $\nu^{N=N}$, $\nu^{N\rightarrow O}$, ν^{C-N} and ν^{N-N}, respectively. The spectrum also shows some bending vibrations at 1515 ± 4, 1052 ± 3, 965 ± 10 cm^{-1}, related to δ^{N-H}, δ^{O-H} and $\delta^{N\rightarrow O}$, respectively.[19,20] The mass spectrum of the each hydroxytriazenes consists of a base peak at m/e =129.7 amu, due to the $[C_6H_3ClF]^+$ fragment. The molecular ion (M^+) appears at m/e M^+ ± 0.5 amu with (M+2) peak. Other fragments observed at m/e = 157.4 ± 0.2 were assigned as $[C_6H_3ClN_2F]^+$ ion.

3-hydroxy-3-phenyl-1-(3-chloro-4-fluorophenyl)triazene (**BPHT**): Mint cream powder; Yield: 81%; m.p. 96 °C; Anal. Calcd. for $C_{12}H_{12}N_3FClO$: C, 64.09; H, 5.63; N, 20.77. Found: C, 63.47, H, 4.98; N, 21.04; IR (KBr cm^{-1}): 3444 (ν^{O-H}), 3221 (ν^{N-H}), 1515 (δ^{N-H}), 1421 ($\nu^{N=N}$), 1332 ($\nu^{N\rightarrow O}$), 1231 (ν^{C-N}), 1200 (ν^{N-N}), 1052 (δ^{O-H}), 956 ($\delta^{N\rightarrow O}$), 1092 (ν^{C-F}), 775 (ν^{C-Cl}). ^{1}H NMR (500 MHz, DMSO-d_6, δ): 12.14(1H, s O···H···N), 7.48(1H, dd, J=4,3, Ar), 7.54-7.59 (3H, m, Ar), 7.64-7.69(1H, dd, J= 2.5&6.5, Ar), 8.11(1H, dd, J=6, Ar), 7.34-7.39(1H, dd, J=2.7&6.2, Ar); MS (m/z, relative abundance,%): 265 (M^+), 128.47(BP).

3-hydroxy-3-(2-methylphenyl)-1-(3-chloro-4-fluorophenyl) triazene **(o-TPHT)**: Yellow crystals; Yield: 67%; m.p. 142 °**C**;Anal. Calcd. for $C_{13}H_{11}ClFN_3O$: C, 55.82; H, 3.96; N, 15.02. Found: C, 55.63; H, 3.88; N, 14.96; IR (KBr, cm^{-1}): 3442 (O-H), 3182 (N-H), 1510 (N-H), 1414 (N=N), 1316 (N→O), 1236 (C-N), 1200 (N-N), 1055 (O-H), 983 (N→O), 1124 (C-F), 771 (C-Cl). ^{1}H NMR (500 MHz, DMSO-d_6, δ): 12.06(1H, s, O···H···N), 7.91(2H, d, J=8, Ar), 7.66 (1H, dd, J=2.5&6.5 Hz, Ar), 7.46-7.49(1H, m, Ar), 7.35-7.40(3H, m, Ar), 2.39(3H, s, $-CH_3$); MS (m/z,): 279 (M^+), 128.76(BP).

3-hydroxy-3-(3-methylphenyl)-1-(3-chloro-4-fluorophenyl) triazene (**m-TPHT)**: Golden Yellow powder; Yield: 87%; m.p. 121 °**C**; Anal. Calcd. for $C_{13}H_{11}ClFN_3O$: C, 55.82; H, 3.96; N, 15.02. Found: C, 56.31; H, 3.84; N, 14.89; IR (KBr, cm^{-1}): 3448 (O-H), 3227 (N-H), 1519 (N-H), 1415 (N=N), 1337 (N→O), 1233 (C-N), 1207 (N-N), 1050 (O-H), 961 (N→O), 1087 (C-F), 776 (C-Cl). ^{1}H NMR (500 MHz, DMSO-d_6, δ): 12.11(1H, s, O···H···N), 7.91(2H, m, Ar), 7.67 (1H, dd, J=2.8&6.5 Hz, Ar), 7.47-7.50(1H, m, Ar), 7.39-7.46(2H, m, Ar), 7.3(1H, d, J=8Hz, Ar), 2.43(3H, s,$-CH_3$); MS (m/z, (relative abundance, %)): 265 (M^+), 68.93 (BP).

3-hydroxy-3-(4-methylphenyl)-1-(3-chloro-4-fluorophenyl) triazene **(p-TPHT)**: Light Yellow crystals; Yield: 82%; m.p. 118 °**C**; Anal. Calcd. for $C_{13}H_{11}ClFN_3O$: C, 55.82; H, 3.96; N, 15.02. Found: C, 55.65; H, 3.86; N, 14.91; IR (KBr, cm^{-}

[1]): 3400 (O-H), 3226 (N-H), 1516 (N-H), 1435 (N=N), 1336 (N→O), 1232 (C-N), 1196 (N-N), 1052 (O-H), 968 (N→O), 1088 (C-F), 775 (C-Cl). ^{1}H NMR (500 MHz, DMSO-d_6, δ): 12.06(1H, s, O···H···N), 7.91(2H, d, J=8, Ar), 7.66 (1H, dd, J=2.5&6.5 Hz, Ar), 7.46-7.49 (1H, m, Ar), 7.35-7.40 (3H, m, Ar), 2.39 (3H, s, -CH_3); MS (m/z, (relative abundance, %)): 265 (M^+), 128.68 (BP).

Co(II)-BPHT: Lion colored powder; Yield: 78%; m.p. 252-254 °**C**; Anal. Calcd. for $C_{26}H_{20}N_6O_2F_2Cl_2Co$: C, 50.62; H, 3.24; N, 13.63. Found: C, 50.22; H, 3.01; N, 13.54.

Co(II)-o-TPHT: Buff colored powder; Yield: 78%; m.p. 204-207 °**C**; Anal. Calcd. for $C_{26}H_{20}N_6O_2F_2Cl_2Co$: C, 50.62; H, 3.24; N, 13.63. Found: C, 50.22; H, 3.01; N, 13.54.

Co(II)-m-TPHT: Buff colored powder; Yield: 69%; m.p. above 250 °**C**; Anal. Calcd. for $C_{26}H_{20}N_6O_2F_2Cl_2Co$: C, 50.62; H, 3.24; N, 13.63. Found: C, 50.39; H; 3.12; N, 13.59.

Co(II)-p-TPHT: Lion colored powder; Yield: 83%; m.p. 190-193 °**C**; Anal. Calcd. for $C_{26}H_{20}N_6O_2F_2Cl_2Co$: C, 50.62; H, 3.24; N, 13.63. Found: C, 50.31; H, 3.08; N, 13.51.

3.2 IR Spectra of complex

IR spectral data of all synthesized Co(II) complexes of hydroxytriazenes are in good agreement with expected range as reported.[20] The bands at 3440, 3200, 1510 and 1050 cm^{-1} corresponding to ν^{O-H}, δ^{N-H} and δ^{O-H} respectively, in the spectrum of all hydroxytriazenes disappeared in the spectra of their Co(II) complexes. This is because, when complexes are formed the hydrogen atom is removed, and therefore the band involving hydrogen must disappear. The $\nu^{N=N}$ and $\nu^{N\rightarrow O}$ bands for the tautomeric triazene 1 oxide [-NH–N=N(→O)-] are of greater intensity in the complexes appearing at 1430 ± 5 and 1330-1346 cm^{-1}, respectively. The strengthening in the bond shows formation of chelate ring by replacing hydrogen atom. The band ν^{C-N} and ν^{N-N} were assigned as 1233 ± 3 and 1200 ± 10 cm^{-1} which are present in all hydroxytriazene were merged to one band indicating delocalization of electrons in chelate rings. The greater value of ν^{N-N} stretching indicates strengthening of N-N bond. The results thus confirms complex formation. The results have included in table-1.

Table 1. *Characteristic IR bands (cm^{-1}), molar conductance and magnetic moment of the cobalt complexes*

Compound	$\nu_{N=N}$	$\nu_{N\rightarrow O}$	ν^{C-N} & ν^{N-N}	$\delta_{N\rightarrow O}$	ν_{C-F}	ν_{C-Cl}	Molar conductance *mho cm^2/ mole*
Co(II)BPHT	1428	1346	1224	1003	1095	752	2.5
Co(II)o-TPHT	1430	1331	1226	989	1061	748	3.2
Co(II)m-TPHT	1432	1338	1232	990	1060	770	6.8
Co(II)p-TPHT	1427	1343	1234	1004	1060	798	4.1

3.3 UV spectra and molar conductivity measurements

The electronic spectra of cobalt complexes in DMSO gave a broad high intensity bend around 27,000 ± 30 cm^{-1} assigned to ligand to metal (L→M) charge transfer. The molar conductance values of the Co(II) complexes in acetonic medium at 10^{-3} M were found in the range of 2.5 to 7 mho cm^2 $mole^{-1}$ (table-1), much less than expected for 1:1 electrolytes[21], hence all complexes are considered as non-electrolytes.

3.4 Magnetic susceptibility measurements

The magnetic moments were measured at room temperature. The observed magnetic moments for the cobalt(II) complexes are in the range 4.37-4.58 BM, indicating tetrahedral environment around Co(II).[22]

3.5 Thermogravmetric analysis

Typical TGA curve of Co(II)p-TPHT complex showed the absence of water molecules, as sudden weight loss was observed at 212 °**C**. The total weight loss was 50.51% (calc. 48.39), which corresponds to a weight loss of molecular weight 298.29 units. Similarly, the weight loss at 234-780 °C was 36.31% (36.25), which corresponds to a weight loss of the remaining organic part to cobalt oxide. Thus TGA curves indicate a 1 : 2 complex in accordance with the analytical data.

3.6 Studies of Complex Formation in Solution: Molar ratio method :

Figure 2 shows typical absorption spectrum for mole ratio method of the complex of hydroxytriazenes with Co(II) ions. The results of study suggest that maximum change of the slope for all the system is observed for the 2 molar ratio of hydroxytriazene ligands for each Co(II) ion corresponding to 1:2 Co(II): hydroxytriazene.

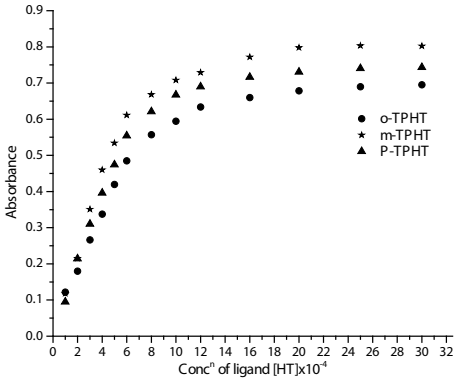


Figure 2. *Mole ratio method*

Continuous variation method:

Figure 3 presents typical absorption spectra for Job's continuous variation method for all the three complexes exhibiting maximum absorbance value at a hydroxytriazene molar fraction close to 1/3. Job's method confirms the Co(II) to hydroxytriazene ligand ratio is 1:2 for each set of complex.

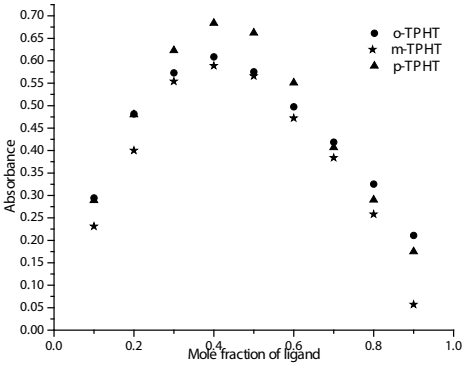


Figure 3 *Job's continuous variation method*

3.7 Antimicrobial screening:

This study was carried out to investigate the antimicrobial activity of hydroxytriazenes and their Co(II) complexes. The m-TPHT showed promising activity in in-vitro models against *C. albicans* (MIC 25µg/mL), *C. neoformans* (MIC 50 µg/mL), *T. mentagrophytes* (MIC 50 µg/mL) and *C. parapsilosis* (MIC 50 µg/mL). Compounds *BPHT* and o-TPHT were found to show good activities against *C. albicans* (MIC 50 µg/mL), *C. neoformans* (MIC 50 µg/mL), *T. mentagrophytes* (MIC 50 µg/mL), *C. parapsilosis* (MIC 50 µg/mL) and *T. mentagrophytes* (MIC 50 µg/mL). In our antimicrobial studies, cobalt complexes excepting Co(*BPHT*) were found to shown good antifungal activity against *C. albicans, C. neoformans, S. schenckii* and *T. mentagrophytes* (MIC 50 µg/mL). Neither hydroxytriazenes nor their cobalt complexes have been found to show antibacterial activity at 50 µg mL^{-1} in *in-vitro* models (Table-2).

Table 2. *Antimicrobial activity of Hydroxytriazenes and their cobalt complexes (MIC's in* **µg/mL***)*

Compounds	Minimum inhibitory conc. (MIC) in against-									
	Bacteria				Fungi					
	A	B	C	D	E	F	G	H	`I	J
BPHT	>50	>50	>50	>50	50	50	>50	50	>50	50
o-TPHT	>50	>50	>50	>50	50	50	>50	50	>50	50
m-TPHT	>50	>50	>50	>50	25	50	>50	50	>50	50
p-TPHT	>50	>50	>50	>50	>50	>50	>50	50	>50	>50
Co(II)BPHT	>50	>50	>50	>50	>50	>50	50	50	>50	>50
Co(II)o-TPHT	>50	>50	>50	>50	50	50	50	50	>50	>50
Co(II)m-TPHT	>50	>50	>50	>50	50	50	50	50	>50	>50
Co(II)p-TPHT	>50	>50	>50	>50	50	50	50	50	>50	>50

Bacteria

A. *E. coli* (ATCC9637),
B. *Pseudomonas aeruginosa,*
C. *Staphylococcus aureus,*
D. *K. Pneumoniae*

Fungi

E. *Candida albicans*
F. *Cryptococcus neoformans*
G. *Sporothrix schenckii*
H. *Trichophyton mentagrophytes*
I. *Aspergillus fumigatus*
J. *Candida parapsilosis* (ATCC-22019).

4. CONCLUSION

We synthesized a series of hydroxytriazenes having chloro fluoro substituents as a bidentate chelating agent. Neutral Co-BPHT, Co(II)o-TPHT, Co(II)m-TPHT and Co(II)p-TPHT complexes were obtained by ligand reactions with cobalt nitrate. The compounds were fully characterized by elemental analyses, FT-IR, 1H NMR, TGA, molar conductance and magnetic susceptibility measurement methods. It has been confirmed by IR that hydroxytriazenes are coordinated to cobalt via hydroxyl and diazo nitrogen. The results show tetrahedral geometry for cobalt complex. The thermogravimetric analyses showed that absence of coordinated water and the complexes decompose in two stages in first partial loss of the organic moiety, and finally the cobalt oxides remained. Results of mole ratio and jobs method has suggested 1:2 stiochiometery of Co(II) and Hydroxytriazenes, respectively. Antimicrobial activities are also reported as minimum inhibitory concentration (MIC) values, defined as the lowest concentration of an antimicrobial that visibly inhibits growth of the bacteria after overnight incubation. These results may be used to develop new compounds involved in analytical, catalytic, bioorganometallic or medical applications.

ACKNOWLEDGEMENTS

The authors are grateful to CDRI Lucknow for providing antimicrobial activity, The SAIF, IIT Madras, Chennai for providing spectral analysis facility and for financial support from CSIR New Delhi

REFERENCES

1. Zabeen R, Goswami AK, Purohit DN, Chim. Acta Turc. 1994, 22, 22.
2. Sharma K, Maroo U, Maroo P, Chauhan RS, Goswami AK, Purohit DN, Asian J. Chem. 1999, 11, 1067.
3. Jain CP, Chauhan RS, Goswami AK, Purohit DN, Asian J. Chem. 2000, 12 909.
4. Paliwal K, Gorji D, Kumar S, Naulkha N, Goswami AK, Purohit DN, Asian J. Chem. 2001, 13, 299.
5. Khan S, Dashora R, Mehta A, Goswami AK, J. Indian Chem. Soc. 2003, 80 793.
6. Naulakha N, Goswami AK, J. Indian Chem. Soc. 2004, 81, 438.
7. Horner M, Giglio VF, Iglesias BA, Martien PR, Michelot TM, Brenner LG, Paraginski GL, Horner R, Brazilian J. Pharm. Scie. 2008, 44, 441.
8. Hura IS, Naulakha N, Goswami AK, Shrivastav MK, Indian J. Microbiology, 2003, 43, 275.
9. Goswami AK, Purohit DN, Anal. Scie. 2001, 17, 789.
10. Jain CP, Chauhan RS, Goswami AK, Bioscie. Biotech. Res. Asia 2006, 3, 381.
11. Singh K, Patel P, Goswami AK, E.J.Chem. 2008, 5, 1144.
12. Chauhan LS, Jain CP, Chauhan RS, Goswami AK, Asian J. Chem. 2007, 19, 4684.
13. Chauhan LS, Jain CP, Chauhan RS, Goswami AK, Adv. Phaemacol. Toxicol, 2006, 7, 73.
14. Chauhan LS, Jain CP, Chauhan RS, Goswami AK, J. Chem. Pharm. Res. 2010, 2, 539.
15. Baroliya PK, Mehta A, Dashora R, Chauhan RS, Goswami AK, Res. Chem. Intermed. 2012, 38, 2149.
16. Rosenberg B, VanCamp L, Krigas T, Nature, 1965, 205, 698.
17. Giaccone G, Drugs, 2000, 59, U4.
18. Purohit DN, Nizamuddin J, J. Indian Chem. Soc. 1983, LX, 712.
19. Purohit DN, Sogani NC, J. Indian Chem. Soc. 1964, 41, 160.
20. Purohit DN, Spectrophotometrica Acta, 1985, 41, 873.
21. Geary WJ, Coord. Chem. Rev. 1971, 7, 81.
22. Figgis BN, Lewis J, In Lewis J, Wilkins RG Eds, Modern Coordination Chemistry; Interscience Publishers Inc. New York, 1960; 400-454

23. Tandon A, Mukerji AK, Verma PS, Mukerji SK, Syn. React. Inorg. Met. 1991, 2, 479.
24. Zeinab H, Abd El-wahab, J. Coord. Chem. 2008, 61, 1696.
25. National Committee for Clinical Laboratory Standard, 1997. Reference method for broth dilution antifungal susceptibility testing of yeast, approved standard. Document M27-A. National Committee for Clinical Laboratory Standards, Wayne, PA, USA.
26. National Committee for Clinical Laboratory Standard, 1998. Reference method for broth dilution antifungal susceptibility testing of conidium forming filamentous fungi: proposed standard. Document M38-P. National Committee for Clinical Laboratory Standard, Wayne, PA, USA.