International Journal of Chemical Science
www.chemicaljournals.com
Online ISSN: 2523-2843, Print ISSN: 2523-6075
Received: 11-05-2021, Accepted: 26-05-2021, Published: 10-06-2021
Volume 5, Issue 3, 2021, Page No. 27-34

Synthesis, physicochemical and spectroscopic analysis of macrocyclic complexes of Fe (III) bearing nitrogen and oxygen as donor atoms and their antimicrobial screening

Sangeeta Lamba, Anita Kumari, Swati Bugalia*
Department of Chemistry, University of Rajasthan, Jaipur, Rajasthan, India

Abstract

In this work, we have synthesized some new oxaazamacrocyclic complexes of iron (III) by metal template condensation synthesis method. [1+1] cyclocondensation reaction of 1,13-diamino,4,7,10-trioxatridecane and various α- diketones such as 2,3-butanedione, 2,3-pentanedione, 3,4-hexanedione,1-phenyl-1,2-propanedione, benzil or 4,4′-dimethylbenzil precursors in presence of $FeCl_3$ [1:1:1] forms N_2O_3-macrocyclic complex of iron (III). The synthesized complexes characterized by elemental analyses, molar conductance, magnetic measurement, electronic, IR, H^1-NMR and mass spectral studies. On the basis of spectral data a six coordinate distorted octahedral geometry has been proposed for all these complexes. Antimicrobial screening of all these complexes have also been carried out by taking some pathogenic bacteria strain *Streptomyces griseus* (gram-positive, MTCC-706), *Escherichia coli* (gram-negative, MTCC-1652), *Staphylococcus aureus* (gram-positive, MTCC-737) and *Pseudomonas aeruginosa* (gram-negative, MTCC-1688) and fungal strain *Trichoderma Reesei* (MTCC-164), *Aspergillus niger* (MTCC-282), *Penicillium funicolusum* (MTCC-1013) and *Fusarium oxysporium* (MTCC-2480). It is observed that Fe(III) complexes show more inhibitory activity than the ligand. All the complexes showed higher activities against *S. griseus* and *E. coli*.

Keywords: N_2O_3 macrocyclic complex, elemental analyses, spectral studies and antimicrobial screening

Introduction

Nowadays, investigation of new complexes having significance in various fields like inorganic synthesis, dyes, catalytic, analytical, industrial, pharmacological, biomedical *etc* is the most important task for the researchers. Macrocyclic complexes have played an important role in the development of coordination chemistry as they readily form stable complexes (by cyclisation) with a variety of metals and now become a major growing area of research because of its applications and importance in industrial, drug, biomedical, polymeric, catalytic etc. fields. Natural and synthetic macrocycles continue to attract extensive scientific interest, with research developments proceeding in many directions including anion receptors, molecular recognition, drug discovery, therapeutics and nanoscience [1, 4]. There are enormous papers reported on synthesis and characterization of macrocyclic complexes using transition metals as central metal ion. Claudel *et al.* [5] reported a review on recent advances and finding of new antimicrobial strategies based on complexes of various metals like iron, zinc, silver, copper, vanadium, ruthenium *etc*. Mekhail *et al* [6] used tetra-aza pyridinophanes ligand with modification to the pyridyl moiety{4-hydroxyl (L1), 4-H (L2), 4-chloro (L3), 4-trifluoromethyl (L4), 4-nitrile (L5), and 4-nitro (L6)}and synthesized six tetra–aza macrocyclic complexes metalated with Fe(III) and characterized by X-ray diffraction, cyclic voltammetry and metal binding affinities (log β). Singh *et al* [7] synthesized a new series of trivalent metal ion macrocyclic complexes of chromium and iron of the type $[M(C_{20}H_{14}N_{10}O_4X]X_2$; by template condensation reaction of oxaldihydrazide and isatin and characterized with the help of elemental analyses, conductance measurements, electronic, IR, Far IR, mass spectral studies. *In vitro* antimicrobial screening of these complexes were also carried out to assess their inhibiting potential. Sharma *et al* [8] studied two newly synthesized tetraaza[N_4] macrocyclic complexes of Fe^{III} and Co^{II} by various analytical techniques IR, UV-Vis, MS and electrochemical behavior was carried out using cyclic voltammmetry on Pt dish electrode. A lot of papers have been reported on macrocyclic complexes of transition metals showing antifungal, antibacterial, anticancer, antiviral, anti-HIV activity, carcinostatic activity, and used as DNA-binding agents, and as useful laboratory and industrial homogenous catalysts [9, 28].

Material and method

Ferric chloride (Merck) of AR grade, 2,3-butanedione (Merck), 2,3-pentanedione (Merck), 3,4-hexanedione (Sigma-Aldrich), 1-phenyl-1,2-propanedione (Sigma-Aldrich), pentane-2,4-dione (Merck), benzil (Merck), 4,4ʹ-dimethylbenzil (Fluka), 1-phenylbutane-1,3-dione (Merck), 1,3-diphenylpropane-1,3-dione (Merck) and 4,7,10-trioxatridecane-1,13-diamine (Merck) were used. Solvents *viz*. n-butanol and dimethyl sulphoxide (DMSO) were distilled before use.

Iron was estimated volumetrically by using diphenylamine as an internal indicator. C, H, and N were carried out using an elemental analyzer FLASH 2000. Molar conductances were measured at room temperature in DMF by a Systronic Direct Reading Conductivity Meter-304 using a glass cell having cell constant 1.0 Cm^-. Magnetic measurements were carried out on Gouy balance calibrated with $Hg[Co(NCS)_4]$. Electronic spectra (DMSO) were recorded in the region 200-800 nm on a Hitachi U-2000 spectrophotometer. The IR spectra were recorded as KBr pellets in the region 4000-400 Cm^- on Shimadzu-8400 S FTIR spectrophotometer. The mass spectra were recorded on WATERS, Q-TOF MICROMASS (LC-MS). ^{1}H- NMR spectra were recorded in DMSO-d$_6$ on JEOL FX 90 QFT NMR spectrometer at 90 MHz using TMS as a reference.

Synthesis of Macrocyclic Complexes of Fe (III)

$FeCl_3$ (0.68 g, 4.2 mmol) was dissolved in ~ 20 ml of n-butanol with stirring in round bottom flask. To this solution 2,3-butanedione (0.36 g, 4.2 mmol) in ~ 15 ml n-butanol was added drop wise with stirring. A butanolic solution of 4,7,10-trioxadiaza-1,13-diamine (0.92 g, 4.2 mmol) added drop wise with a constant stirring. The pH of solution was 3.5. Add 10% NaOH solution drop wise to the reaction mixture to raise the pH up to 7. The precipitate began to appear. The contents were stirred for ~ 5 hrs. The resulting solid was filtered, washed with petroleum ether and dried under reduced pressure. The characteristics and analyses of the resulting complexes are mentioned in Table 1.

Table 1: Physical perameters of trioxa-diazamacrocycles complexes of Fe(III).

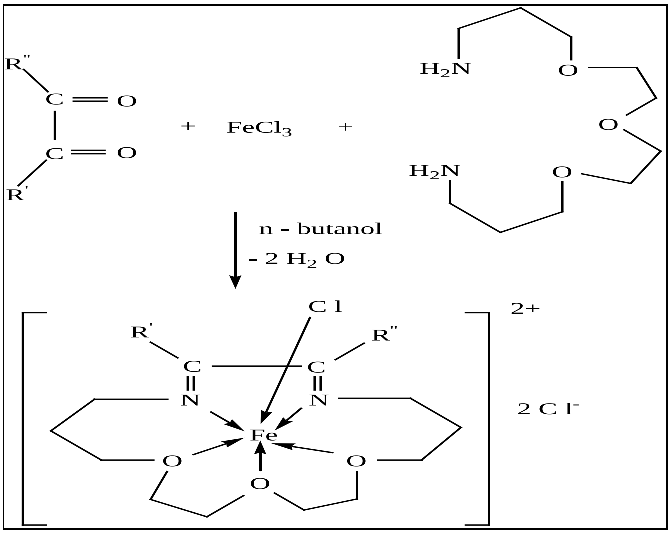
S. No.	Complex Formula Weight(g)	Color Decomp. Temp. (℃)	Yield (%)	Found (Calculated) %			
				Fe	C	H	N
1.	$[Fe(C_{14}H_{26}O_3N_2)Cl)]Cl_2$ 432.57	Brown (240)	55	12.80 (12.91)	38.90 (38.87)	6.30 (6.06)	6.20 (6.48)
2.	$[Fe(C_{15}H_{28}O_3N_2)Cl)]Cl_2$ 446.60	Brown (237)	62	12.30 (12.50)	40.50 (40.34)	6.25 (6.31)	6.30 (6.27)
3.	$[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl_2$ 460.63	Brown (244)	58	12.00 (12.12)	41.60 (41.72)	6.55 (6.56)	6.05 (6.08)
4.	$[Fe(C_{19}H_{28}O_3N_2)Cl)]Cl_2$ 494.64	Brown (218)	60	11.20 (11.29)	46.20 (46.14)	5.60 (5.71)	5.72 (5.66)
5.	$[Fe(C_{24}H_{30}O_3N_2)Cl)]Cl_2$ 556.70	Brown (235)	49	9.98 (10.03)	51.60 (51.78)	5.45 (5.43)	5.20 (5.03)
6.	$[Fe(C_{26}H_{34}O_3N_2)Cl)]Cl_2$ 584.75	Brown (234)	57	9.60 (9.55)	53.45 (53.40)	5.70 (5.86)	5.00 (4.79)

Pharmacology

Clinical laboratory bacterial isolates of *Streptomyces griseus* (gram +ve, MTCC-706), *Escherichia coli* (gram -ve, MTCC-1652), *Staphylococcus aureus* (gram +ve, MTCC-737) and *Pseudomonas aeruginosa* (gram -ve, MTCC-1688) were used for antibacterial studies and fungal isolates of *Trichoderma Reesei* (MTCC-164), *Aspergillus niger* (MTCC-282), *Penicilliumfunicolusum* (MTCC-1013) and *Fusarium oxysporium* (MTCC-2480) were screened for evaluation antifungal studies of the synthesized macrocyclic complexes.Mueller Hinton Agar (for bacteria) and Sabouraud’s dextrose agar, SDA (Merck) (for fungi) were used for the biological assay. Both antibacterial and antifungal assessment of the samples was carried by the agar well diffusion method.

Result and Discussion

The reaction of α-diketones such as 2,3-butanedione, 2,3-pentanedione, 3,4-hexanedione, 1-phenyl-1,2-propanedione, benzil or 4,4′-dimethylbenzil and 4,7,10-trioxatridecane-1,13-diamine with $FeCl_3$ in (1:1:1) molar ratios have resulted the formation of Fe(III) complexes of 17-membered N_2O_3 macrocycles (Scheme 1).



R'	R''
CH_3	CH_3
CH_3	C_2H_5
C_2H_5	C_2H_5
CH_3	C_6H_5
C_6H_5	C_6H_5
p-$CH_3C_6H_4$	p-$CH_3C_6H_4$

Scheme 1: Synthesis of Fe (III) trioxa-diazamacrocyclic complexes derived from diamine and α-diketones

The resulting macrocyclic complexes are brown colored solids. These complexes are quite stable at room temperature but decompose at high temperature. All the complexes are insoluble in water, carbon tetrachloride, methanol, chloroform and ethanol but soluble in dimethylsulphoxide and dimethylformamide. The percentage of Fe, C, H, and N are compatible with calculated values.

Molar Conductance

The molar conductance values for all the macrocycles of Fe(III) (10^{-3} M) were determined in DMSO. These values were found in the range of 125-152 ohm^{-} cm^{2} mol^{-} indicating 1:2 electrolytic nature (Table 2).

Magnetic Moments

The magnetic moments (μ_{eff}) for the iron(III) macrocyclic complexes observed in the range 5.78 to 5.92 μ_B indicating their paramagnetic behavior (Table 2). This corresponds to five unpaired electrons typical of the d^5 system [29].

Table 2: Molar conductances and magnetic moment of Fe(III) complexes

S. No.	Complex	Conductances*(Ohm^{-1}cm^{2}mol^{-1})	Magnetic moment(μ_B)
1.	$[Fe(C_{14}H_{26}O_3N_2)Cl)]Cl_2$	138	5.82
2.	$[Fe(C_{15}H_{28}O_3N_2)Cl)]Cl_2$	125	5.89
3.	$[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl_2$	134	5.92
4.	$[Fe(C_{19}H_{28}O_3N_2)Cl)]Cl_2$	152	5.86
5.	$[Fe(C_{24}H_{30}O_3N_2)Cl)]Cl_2$	148	5.84
6.	$[Fe(C_{26}H_{34}O_3N_2)Cl)]Cl_2$	130	5.78

Infrared Spectra

A band observed at 3200-3400 cm^{-1} region in the spectra of 4,7,10-trioxatridecane-1,13-diamine, assigned to ʋ(NH$_2$). In the infra-red spectrum of iron(III) complexes, these bands {ʋ(C=O) and ʋ(NH$_2$)} are disappear and a new medium intensity band in 1585-1630 cm^{-1} confirm the condensation of >C=O group of α-diketones and –NH$_2$ group of oxa-azadiamine and formation of macrocyclic schiff's base. These bands may be assigned due to ʋ(C=N) stretching vibrations. Cummings and Busch observed the absence of ʋ(NH$_2$) and ʋ(C=O) frequencies in their research work[30]. A weak to medium intensity absorption bands in the region 430-550 cm^{-1}, which are absent in free ligands, may be assigned as ʋ(Fe-O) vibrations. Shelke and coworkers have reported a band in the region 457-540 cm^{-} for Cu(II), Co(II), Mn(II), La(III) and Ce(III) complexes derived from bidentate Schiff base, N-benzylidene-2-hydroxybenzohydrazide[31]. All Fe(III) macroccyclic complexes exhibit absorption bands in the region 2830-2915 cm^{-1} which can be assigned to aliphatic υ(C-H) stretching vibration.

Panchbhai *et al.* have reported υ(C-H) stretching and υ(C-H) bending absorption bands in the region 2850-2890 cm^{-1} and 1410-1480 cm^{-1} respectively for Ru(II), Pt(II) and Pd(II) complexes of macrocyclic ligand 5,12-dioxa-7,14-dimethyl-1,4,8,11-tetraazacyclotetra-deca-1,8-diene[32]. In the IR spectra of macrocyclic complexes derived from 1-phenyl-1,2-propanedione, benzil or 4,4'-dimethylbenzil, a medium intensity absorption band in the region 1410-1500 cm^{-1} may be assigned for phenyl ring absorption.The spectrum of $[Fe(C_{14}H_{26}O_3N_2)Cl]Cl_2$ complex is reproduced in Figure.1.

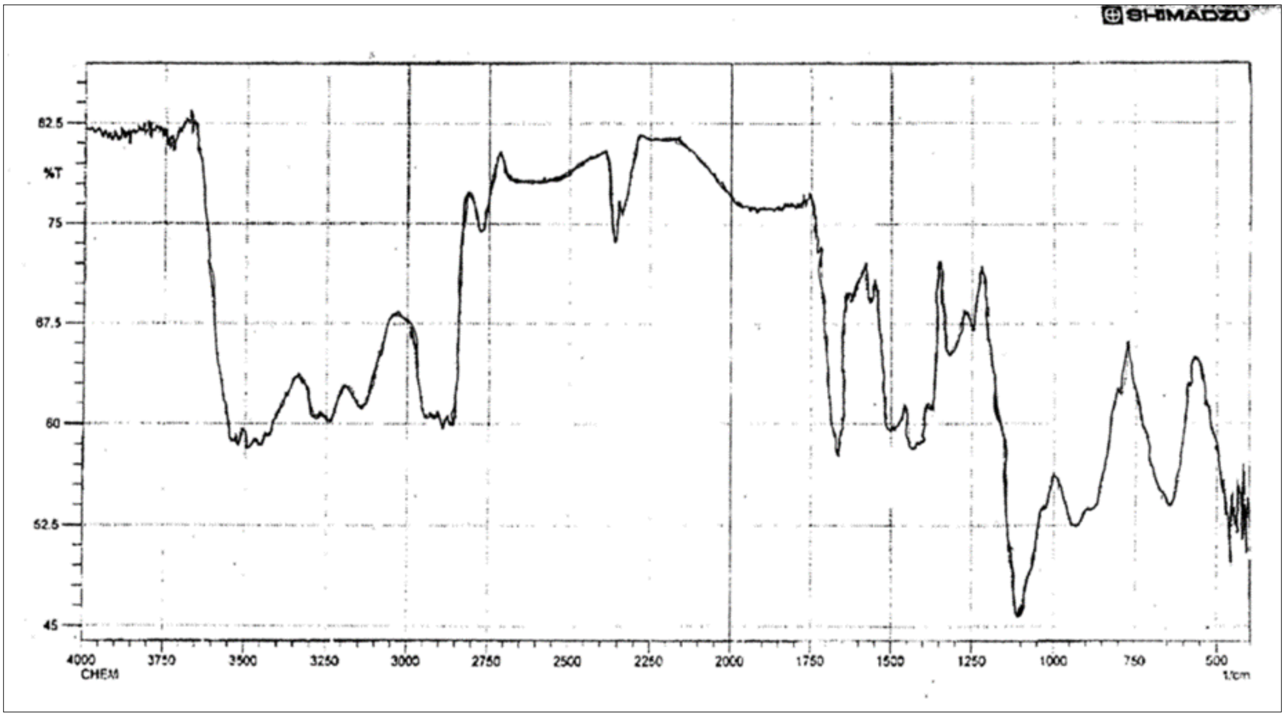


Fig 1: IR Spectrum of $[Fe(C_{14}H_{26}O_3N_2)Cl]Cl_2$

Electronic Spectra

Fe(III) has d^5 (t_{2g}^3 e_g^2) high spin configuration in octahedral complexes. The ground state term for d^5 Fe(III) is $^6A_{1g}$. Following three types of transitions are possible: $^6A_{1g} \rightarrow {}^4T_{1g}$, $^6A_{1g} \rightarrow {}^4T_{2g}$, and $^6A_{1g} \rightarrow {}^4A_{1g}$. The electronic spectra of Fe(III) macrocyclic complexes of oxa-azamacrocycles exhibit bands in the region 12955-13370 cm^{-1}, 21525-22110 cm^{-1}, 26750-27225 cm^{-1} assigned to $^6A_{1g} \rightarrow {}^4T_{1g}$, $^6A_{1g} \rightarrow {}^4T_{2g}$ and $^6A_{1g} \rightarrow {}^4A_{1g}$ transitions, respectively. Shirode reported bands in the range 12500-12529, 21270-21296 and

26978-27032 cm^{-1} assignable to $^6A_{1g} \rightarrow {}^4T_{1g}$ (G), $^6A_{1g} \rightarrow {}^4T_{2g}$ (G) and $^6A_{1g} \rightarrow {}^4A_{1g}$ (G) transitions for Fe(III) complexes of the formulae $[Fe(APS)(AS)(H_2O)_2]Cl_3$ synthesized by the reaction of metal chloride with acetophenone semicarbazone and acetone semicarbazone, which indicates the octahedral geometry around iron [33].

Table 3: Absorption bands in the electronic spectra of Fe(III) complexes

S. No.	Complex	$^6A_{1g} \rightarrow {}^4T_{1g}$ (cm^{-})	$^6A_{1g} \rightarrow {}^4T_{2g}$ (cm^{-})	$^6A_{1g} \rightarrow {}^4A_{1g}$ (cm^{-})
1.	$[Fe(C_{14}H_{26}O_3N_2)Cl)]Cl_2$	12990	21780	26750
2.	$[Fe(C_{15}H_{28}O_3N_2)Cl)]Cl_2$	13250	21525	27145
3.	$[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl_2$	12955	21875	26860
4.	$[Fe(C_{19}H_{28}O_3N_2)Cl)]Cl_2$	13175	22105	27225
5.	$[Fe(C_{24}H_{30}O_3N_2)Cl)]Cl_2$	13335	22110	26965
6.	$[Fe(C_{26}H_{34}O_3N_2)Cl)]Cl_2$	13370	21985	26990

^{1}H NMR

Proton nuclear magnetic resonance spectra were recorded in DMSO-d_6 at 90 MHz by using TMS as internal reference and the δ values (ppm) for different protons of macrocyclic complexes of Fe(III) are given in Table 4. A peak in the region δ 2.48-2.59 ppm is observed in the spectra of all the macrocyclic complexes due to the residual methyl protons of the solvent DMSO-d_6. A peak in free diamine due to the -NH_2 at δ 1.48 ppm disappears in the spectra of complexes confirm the condensation of -NH_2 group of diamine with >C=O group of ketone to give macrocycles having >C=N linkages. Similar kinds of results were reported by Prasad *et al.* in 17-membered N_2O_3 macrocyclic complexes of Mg(II), Ca(II), Sr(II) and Ba(II) derived by the [1+1] cyclocondensation of 1,13-diamino-4,7,10-trioxatridecane with α-diketones such as benzil or 4,4′-dimethylbenzil in the presence of alkaline earth metal ions as templates [34].

Table 4: Chemical Shifts (δ,ppm) of the protons in ^{1}H NMR spectra of Fe(III) complexes

S. No.	Complex	Chemical Shifts (δ,ppm)							
		Amine residue					Ketone residue		
		NH_2	NCH_2^a	CH_2^b	-OCH_2^c	-O-CH_2^d-CH_2^d-O-	CH_3^e	-CH_2^f	-CH_3^g
1.	4,7,10-trioxadiaza-1,13-diamine	1.48S	2.55t	1.67m	3.15t	3.45-3.48m	-	-	-
2.	2,3-Pantanedione	-	-	-	-	-	2.27s	2.35q	1.19t
3.	$[Fe(C_{15}H_{28}O_3N_2)Cl)]Cl_2$	-	2.68t	1.74M	3.22t	3.57 – 3.60m	2.04s	2.15m	0.98t

Mass Spectra

The LC-MS mass spectra of the macrocyclic complexes of Fe(III) have been recorded and m/z values and % relative abundances of various fragments of $[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl_2$ complex has been given in Table 5. All the macrocyclic complexes show peaks corresponding to molecular ion $[M]^+$ and free macrocycle $[L]^+$ which confirm the formation of macrocyclic complexes. In addition to the peaks due to the molecular ions and the macrocycles, the spectra exhibit peaks assignable to various fragments arising from the thermal cleavage of the macrocycles and their complexes. The peaks are very useful tool for the determination of structure of a complex.

Table 5: m/z Values and % relative intensities of important peaks in Mass spectrum of $[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl_2$ complex

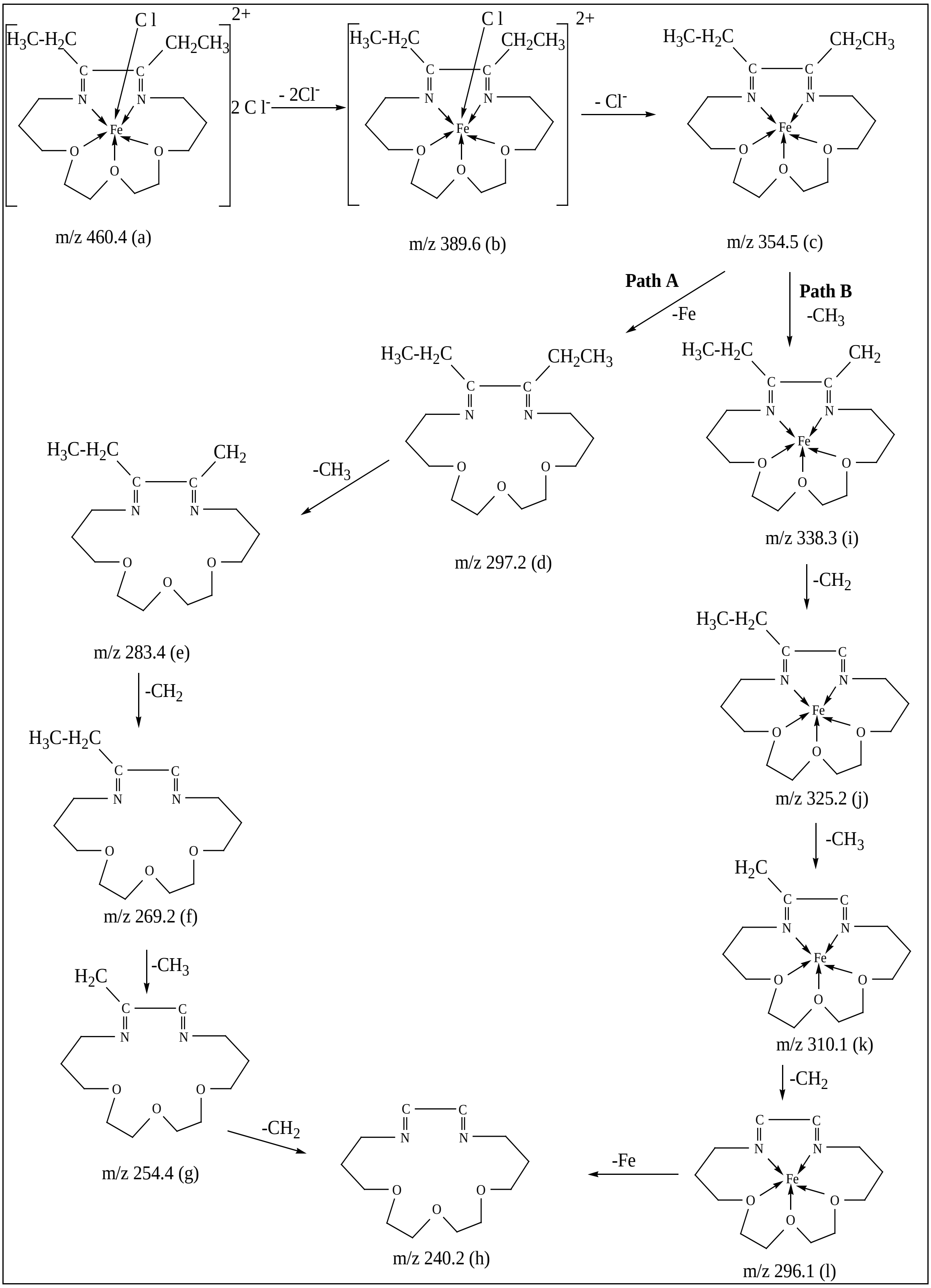
S. No.	Fragments	$[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl_2$ m/z Values (Relative intensities %)
1.	$[M]^+$	460.4 (7%)
2.	$[M + H]^+$	461.4 (5%)
3.	$[M – 2Cl]^+$	389.6 (18%)
4.	$[M – 3Cl]^+$	354.5 (45%)
5.	$[L]^+$	297.2 (8%)
6.	$[L + H]^+$	298.2 (4%)
7.	$[L – CH_3]^+$	283.4 (13%)
8.	$[L – CH_2 – CH_3]^+$	269.2 (4%)
9.	$[L – CH_2–2CH_3]^+$	254.4 (5%)
10.	$[L – 2(CH_2–CH_3)]^+$	240.2 (16%)
11.	$[M– 3Cl – CH_3]^+$	338.3 (9%)
12.	$[M– 3Cl – CH_2 –CH_3]^+$	325.2 (4%)
13.	$[M– 3Cl – CH_2 – 2CH_3]^+$	310.5 (3%)
14.	$[M – 3Cl – 2(CH_2–CH_3)]^+$	297.2 (6%)

The fragmentation pattern of the Fe(III) complex $[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl_2$ is shown in Scheme 2. This complex exhibits molecular ion peak $[M]^+$ (a) at m/z 460.4 (7%). Fragment (b) is produced due to loss of two chloride ion from molecular ion (a) and appears at m/z 389.6 (18%). By the loss of another chloride ion, peak due to the fragment (c) is observed at m/z 354.5 (45%). There are two possible pathways for the fragmentation of (c).

The spectrum of $[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl_2$ has been analyzed on the basis of isotopic abundances of Cl [^{35}Cl (75.78%) and ^{37}Cl (24.22%)] (Table 6). For the molecular ion M^+ isotopic peaks appears at m/z 459.2, 461.2, 463.2 and 465.2. Due to fragment $[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl$ isotopic peaks appears at m/z 424.6, 426.6 and 428.6. Isotopic peaks of fragment $[Fe(C_{16}H_{30}O_3N_2)Cl)]$ appears at m/z 389.2 and 391.2.

Table 6: m/z Values isotopic peaks in Mass spectrum of $[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl_2$ complex

S. No.	Fragment ion composition	m/z
1.	$C_{16}H_{30}O_3N_2Fe^{35}Cl^{35}Cl\ ^{35}Cl$	459.2
2.	$C_{16}H_{30}O_3N_2Fe^{35}Cl^{35}Cl\ ^{37}Cl$	461.2
3.	$C_{16}H_{30}O_3N_2Fe^{35}Cl^{37}Cl\ ^{37}Cl$	463.5
4.	$C_{16}H_{30}O_3N_2Fe^{37}Cl^{37}Cl\ ^{37}Cl$	465.2
5.	$C_{16}H_{30}O_3N_2Fe^{35}Cl^{35}Cl$	424.6
6.	$C_{16}H_{30}O_3N_2Fe^{35}Cl^{37}Cl$	426.6
7.	$C_{16}H_{30}O_3N_2Fe^{37}Cl^{37}Cl$	428.6
8.	$C_{16}H_{30}O_3N_2Fe^{35}Cl$	389.2
9.	$C_{16}H_{30}O_3N_2Fe^{37}Cl$	391.2



Scheme 2: Fragmentation pattern of $[Fe(C_{16}H_{30}O_3N_2)Cl)]Cl_2$

Antimicrobial Screening

All synthesized macrocyclic complexes of Fe(III) were screened against pathogenic fungal and bacterial strains. Antibacterial activity was tested against bacterial strains *viz. Streptomyces griseus* (gram-positive, MTCC-706), *Escherichia coli* (gram-negative, MTCC-1652), *Staphylococcus aureus* (gram-positive, MTCC-737) and *Pseudomonas aeruginosa* (gram-negative, MTCC-1688) while antifungal activity was tested against four fungal pathogens *viz. Trichoderma Reesei* (MTCC-164), *Aspergillus niger* (MTCC-282), *Penicillium funicolusum* (MTCC-1013) and *Fusarium oxysporium* (MTCC-2480) and compared with comparing standard known antibiotics such as Streptomycin and Ketakenazole respectively. Antimicrobial screening shows that the metal complexes exhibit antimicrobial properties against both fungal and bacterial strains. It is important to note that these complexes exhibit more inhibitory effects towards both of the bacterial as well as fungal strains as compare to the parent ligand. The increased activities of the metal complexes as compare to ligand can be explained on the basis of chelation theory. Chelation may enhance the biochemical potency of bioactive species. It enhances the penetration of complex in to lipid membrane and blocking of the metal binding sites in the enzymes of microorganisms. On chelation, the polarity of the metal ion reduced to a great extent due to the partial sharing of its positive charge with donor atoms of ligand and stability of the complex increases by the π-electron delocalization over the whole chelate ring. This process increases the liophilic nature of the compound which favours the penetration of complexes through the cell membrane of microorganisms. These complexes also disturb the respiration process of the cell and thus block the synthesis of proteins, which restricts further growth of the organism [35]. At lower concentration, inhibition is less severe, the activities of the organism will only be slowed down and it may be able to grow at a slow rate, while at higher concentration, more enzymes will become inhibited leading to a quicker death of the organism. Presence of aromatic group increases its inhibitory effect on one or more type of bacteria and fungi as compare to alkyl group in the same position [36].

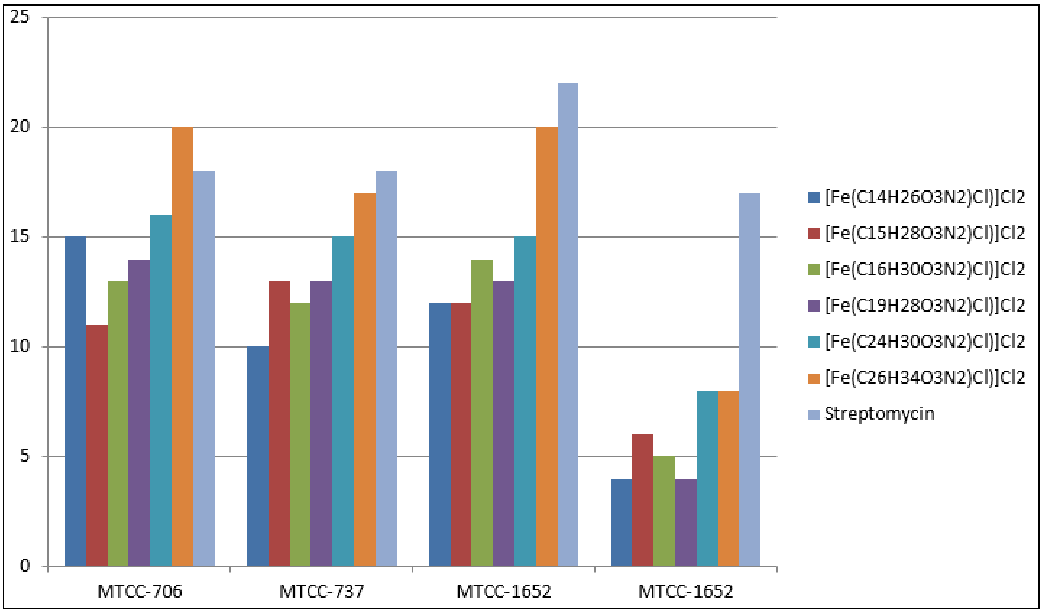


Fig 2: Comparision of MIC (in 100 µg/ml) of macrocyclic complexes and streptomycin against different bacterial strain

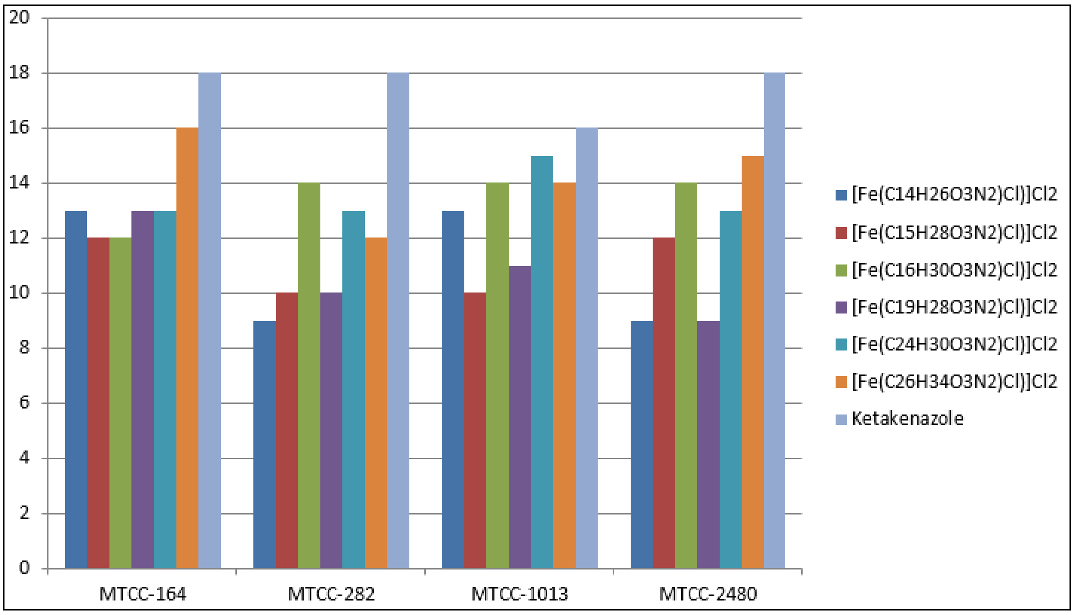


Fig 3: Comparision of MIC (in 100 µg/ml) of macrocyclic complexes and ketakenazole against different fungal strain

Conclusion

In the present work metal complexes were synthesized successfully. On the basis of elemental analyses, molar conductance, magnetic measurement, ^{1}H NMR, IR, UV-Visible and mass spectral studies, a hexa-coordinated environment around the iron metal ion is proposed for all the complexes. The antimicrobial screening of synthesized complexes carried out against the bacterial strains *Streptomyces griesus*, *Escherichia coli*, *Staphylococcus aureus*, *Pseudomonas aeruginosa* and fungal strains Trichoderma *Reesei*, *Aspergillus niger*, *Penicillium funicolusum*, *Fusarium oxysporium*. It is observed that Fe(III) complexes show more inhibitory activity as compared to individual ligands.

Acknowledgement

We are thankful to Head, Department of Chemistry, University of Rajasthan, Jaipur for providing IR spectral studies, Head, Banasthali Vidyapith, Banasthali, Rajasthan for magnetic measurement, Head, SAIF Panjab University, Chandigarh, Punjab for C, H, N analyses and mass spectral studies and Director, Seminal Applied Science Jaipur for antimicrobial screening.

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