

## Complex Formation of Zn(II) Perchlorate Ions Using a Ligand Having Tetraoxotetrahydrazin Moity on Some Pathogenic Bacteria.

M.R.Ullah, M.J.Hossain,  
Department of Chemistry, BUET, Dhaka.

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**Abstract:** Six new macrocyclic complexes were synthesized by template reaction of malanodihydrazide with Zn (II) perchlorate. The metal perchlorate to ligand proper molar ratio and containing a ligand having tetraoxotetrahydrazin moity are synthesized by template condensation of malonodihydrazide ( $C_3H_8N_4O_2$ ) with different aldehydes. The complexes are characterized on the basis of elemental analysis, UV-visible & IR spectroscopy, T.G.A. magnetic moment and conductance measurement and other physical properties. The IR spectrum study of the complex compounds suggests that ligand coordinates to metal ions through the nitrogen atoms from the tetraoxotetrahydrazin moity. Antibacterial activity of the derived complex compounds, as well as already used standard compound kanamycin, was tested on fourteen pathogenic bacteria. Given results were then compared to the efficacy of the Antibacterial activity of standard compound kanamycin used for control of these pathogenic bacteria.

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### I. Introduction

The synthesis and characterization of Schiff base chelates have been interested chemists<sup>(1-2)</sup>. Complexes of some chemistry of synthetic macrocyclic ligands can be divided into two transition metal ions with Schiff base derived from the coupling of difference aldehyde. Firstly there are the cyclic polyether of the 'crown' type of which is a typical example<sup>1</sup>. Ligands of this general category have received much recent attention because of their unusual behavior towards a range of non-transition metal ions<sup>2</sup>. Few studies involving transition metal ions have been reported<sup>3</sup> and it is evident that the majority of such polyether ligands show a limited tendency to form stable complexes with these ions<sup>4</sup>. The coordination chemistry of hydrazones is an intensive area of study and numerous transition metal complexes of these ligands have been investigated<sup>5</sup>. The development of the field of bioinorganic chemistry has increased the interest in Schiff base complexes, since it has been recognized that many of these complexes may serve as models for biologically important species<sup>6</sup>. Coordination compounds derived from aroylhydrazones have been reported because of their anti-tuberculosis, antimicrobial and corrosion inhibitors<sup>7</sup>. The chemistry and complexation properties of macrocyclic dioxotetraamines were investigated<sup>8</sup>. These macrocycles contain two amino nitrogens and two amides. As with cyclam and cyclen, the amino nitrogens with additional coordinating groups form new hexadentate ligands. They are able to bind to metals Zn(II) with simultaneous dissociation of the two amide protons, such that metal binding is highly pH-sensitive and reversible (-a very useful property for metal-sensing applications). The copper(II) complex of a functionalised trans system at neutral and basic pH, and found very different structures according to whether just one or both of the amides are deprotonated<sup>9</sup>.

The recent article describing the use of [Cu-perchlorate contain complex] as a color indicator for solvent parameters<sup>10</sup> fails to identify the potential danger associated with the preparation and handling of this salt. Most of us are aware that "organic perchlorates are self-contained explosives"<sup>11</sup> However, many overlook the fact that a perchlorate salt of a cation, such as a complex ion that contains an organic group or other oxidizable atoms, is also an explosive (although the conditions required to initiate an explosion vary from sample to sample). For example, one sample of  $Co(H_2O)_3(ClO_4)_2$  detonated under a slight impact while attempts to repeat the detonation with other samples were not successful<sup>12</sup>. Such compounds must be handled with great care<sup>13</sup>, if at all.

### Biological Activity of Some Important Compounds

The last decade or so there has been a growing awareness of the importance of wide range of metallic and non metallic elements in biological system<sup>16</sup>. Some 25 elements which are currently throughout to be essential to life, ten can be classified as trace metal ions; Fe, Cu, Zn, Mn, Co, Cr, Sn, V and Ni and four as bulk metal ions; Na, K, Mg and Ca. In addition there is some tentative evidence that Cd and Pb may be required at very low levels. There is also evidence that Sn, As and Br may possibly be essential trace elements. In the following section the out line of the chemistry and biological effects of some of the essential and polluting elements is given bellow.

A number of metal complexes and ligand have been shown to be chemically useful in variety of areas, e.g. As antitumor agent's antiviral agents and in the treatment of illness, for example, in haemocyanins, contain Cu and bind one molecule of O<sub>2</sub> for every pair of copper(I) ions. Haemocyanine is found only in molluscs and arthropods. Inorganic chemistry has been interested in developing suitable copper complexes which would mimic some of properties of haemocyanin.<sup>17</sup>

### **Aim of the Present Work**

In the recent years considerable attention have been given to the synthesis of macrocyclic complex<sup>18</sup>. These complex compounds have been used an model system of biologically important materials, such as porphyrin and corins. Some of the macrocyclic ligand cannot be easily prepared from the reactants<sup>19</sup>. In that case the complex compounds could be synthesised by template method. The desire macrocyclic ligand can be isolated by stripping the complex compounds<sup>20</sup>. Macrocyclic tetraaza complex of Ni<sup>2+</sup> act as catalyst to reduce CO<sub>2</sub> to CO and Fe<sup>2+</sup>, Mn<sup>3+</sup> porphyrins have been most commonly studied catalyst.<sup>21</sup> In view of the extensive use as drugs and significant pharmacological activities of macrocyclic complexes and their derivatives, it is desired to synthesize macrocyclic complexes of Ni (II), Cu (II) and Fe (II). The synthesized macrocyclic complexes and their derivatives are expected to have microbial activity. Therefore, considering the rapid increasing importance of macrocyclic ligand and their complexes in biology and in medicine the present work is divided in to two parts:

1. Firstly, synthesis of some new macrocyclic complexes by the reactions of malonodihydrazide with Zn(II) perchlorate in the presence of formaldehyde, acetaldehyde, butyraldehyde will be characterised by elemental analysis, UV visible and IR spectral analysis, magnetic moment and conductance measurements and some other physical properties.
2. Secondly, study of antibacterial activity of the synthesised complexes (some test organisms such as, *Salmonella-17*, *Klebsilla*, *Shigella dysenteriae*, *Shigella shiga*, *Shigella boydii*, *Shigella sonnei*, *Shigella flexneri*, *Escherichia coli*, *Pseudomonas aeruginosa*, *Salmonella*, *Bacillus megaterium*, *Sarcina lutea*, *Staphylococcus aureus*, *Bacillus cereus*) including the investigation of minimum inhibitory concentration of the complexes.

## **II. Experimental**

The ligand precursor, malonodihydrazide was prepared by the literature procedure<sup>22</sup>. Micro analysis for carbon hydrogen and nitrogen were obtained by using Kjeldahl Method for elemental analysis. Infrared spectra (as KBr disc) were recorded using a shimadzu FTIR-8400 spectrometer from 4000-400 cm<sup>-1</sup> and uv-visible spectra on shimadzu uv-160 Spectrophotometer in DMSO. Magnetic moment measurements were done on Sherwood scientific magnetic susceptibility balance. Conductivities were measured by CG-857 Scott Gerate GmbH conductivity meter with a dip type cell having platinum electrodes in DMSO. Metals were estimated complex complex metrically using EDTA and DMG after fuming the complexes with sulfuric acid<sup>22</sup>. Melting points were determined on an electro thermal melting point apparatus (model no. AZ 6512).

### **Preparation of Macrocyclic Complexes.**

#### **Preparation of [Zn(C<sub>8</sub>H<sub>16</sub>N<sub>8</sub>O<sub>4</sub>)(ClO<sub>4</sub>)<sub>2</sub>] Complex 1.**

To the aqueous malonodihydrazide, C<sub>3</sub>H<sub>8</sub>N<sub>4</sub>O<sub>2</sub> (0.792 g, 6 mmol in 10 mL water) formaldehyde solution (0.48 g, 6 mmol 37%) was added. To the above solution Zinc(II) perchlorate hexahydrate (1.656 g, 3 mmol in 10 mL water) was added and the whole mixture was refluxed with constant stirring for two hours and cooled down. A blue precipitate was formed immediately. The product was washed with ethanol for three times and dried in a vacuum desiccator over anhydrous CaCl<sub>2</sub>. The melting point of the compound was 225<sup>o</sup>C and yield was 2.342 g (80%). The compound was soluble in DMSO and insoluble in acetone, ethanol, water and chloroform. Same procedure was applied for the preparation of complexes 2, 3, 4, 5 and 6 using acetaldehyde, crotonaldehyde, cinnamaldehyde, benzaldehyde and butanal dehyde were the reaction mixture was refluxed for 4, 5, 3, 7 and 10 hours respectively.,

## **III. Result And Discussion Macrocyclic Complexes Of Zn (II)**

Reactions of malonodihydrazide with Zn(II) perchlorate hexahydrate in presence of formaldehyde, acetaldehyde, crotonaldehyde, cinnamaldehyde, benzaldehyde and butanal dehyde give some 16 member macrocyclic complex as described above Complexes (1-6) are characterized on the basis of elemental analysis, magnetic moment & conductance measurements, UV-visible spectra & infrared studies and other physical properties, like melting point, solubility, colour etc. Molar conductance data of the complexes (1-6) are shown in Table 4.1. The conductance values of the complexes suggested that they are non-electrolytic in nature<sup>24</sup>. The infrared spectra of the complexes (1-6) are shown as spectral data (Table 4.4) of the complexes showed a strong and broad band at (3246-3265) cm<sup>-1</sup> which is assigned for the ν(NH) stretching<sup>25</sup>.

Due to coordination the  $\nu(\text{N-H})$  stretching of the amide group goes to the higher field at (3246, 3265)  $\text{cm}^{-1}$  region as compared to the starting material malonodihydrazide<sup>26</sup>. In the complexes the terminal- $\text{NH}_2$  group of malonodihydrazide condensed with the aldehyde moiety form a new secondary<sup>1</sup>-NH group which may appear at the same region (or overlape) as to the amide-NH group as a result the  $\nu(\text{N-H})$  band appear as a strong and broad band. [The starting material malonodihydrazide have three  $\nu(\text{N-H})$  bands at (3248, 3213, 3050)  $\text{cm}^{-1}$ . The bands at (3248, 3050)  $\text{cm}^{-1}$  for the asymmetric and symmetric  $\nu(\text{N-H})$  stretching of the terminal- $\text{NH}_2$  moiety and 3213  $\text{cm}^{-1}$  for amidic (N-H) group]. The complexes showed a broad band at (2920-2972)  $\text{cm}^{-1}$  is suggested for the  $\nu(\text{C-H})$  stretching of aliphatic moiety<sup>32</sup>. The complexes showed a strong band at (1649-1674)  $\text{cm}^{-1}$  which represent the  $\nu(\text{C=O})$  of NH-NH-CO- $\text{CH}_2$  moiety<sup>27</sup>. Three or four band at (625-1145)  $\text{cm}^{-1}$  region also indicated the  $\nu_1, \nu_2, \nu_3, \nu_4$  bands of ( $\text{ClO}_4^-$ ) moiety. These stretching frequency is suggested the coordination of perchlorate to the metal through the O atom<sup>28</sup>. A medium band at (407-412)  $\text{cm}^{-1}$  region is tentatively attributed to the  $\nu(\text{M-N})$  mode<sup>29,30</sup>. indicating the coordination of the ligand to the metal through the nitrogen atom. The magnetic moment measurement data (Table 4.3) of the Zn(II) complexes (1-6) showed (1.56-1.78) B.M. These values correspond to no unpaired electrons of Pb(II)  $d^{10}$  system suggest the octahedral environment of the complexes which are consistent with the literature value<sup>1</sup>. The elemental analyses (C, H and N) (Table 4.2) and metal estimation data (Table 4.3) of the complexes are consistent with the proposed formula. The UV-visible spectra of the complexes (1-6) are shown (Table 4.5) band at 320,480 nm, (1-6) at represent the d-d transition of  $^4A_{2g}(\text{F}) \rightarrow ^4T_{1g}(\text{F}), ^4A_{2g}(\text{F}) \rightarrow ^4T_{1g}(\text{P})$ , which suggested the octahedral geometry of the Pb (II) complexes<sup>31,32</sup>.

**Thermal studies:** The thermal properties of metal (II) complexes were investigated by thermograms (TGA,DTA) and are shown in (Fig 4.2) and the corresponding thermal analysis is presented in (Tabl.4.6). In the case of complex (I) (Fig. 01) the decomposition occurs in the (230-325)<sup>0</sup>C range. There is no mass loss up to 230<sup>0</sup>C. The first stage of decomposition starts at 230<sup>0</sup>C and end at 230<sup>0</sup>C with a corresponding weight loss 25%. Which is accompanied by endothermic effect in the DTA curve in the range 225<sup>0</sup>C which is accompanied by weight loss confirming the second stage of decomposition is observed at 225-350<sup>0</sup>C (60% wt. loss). meanwhile the DTA curve exhibits endothermic effect in the range 325<sup>0</sup>C which is accompanied by weight loss confirming. On the basis of elemental analysis magnetic moment and conductance measurements, thermal studies UV Visible spectra, infrared spectra and other physical properties the suggested structure of the complexes are octahedral in nature as in Fig.4.1.

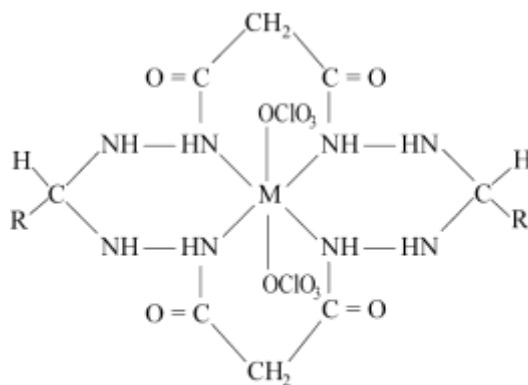


Fig. 4.1

M =Zn(II), where R=H(1), CH<sub>3</sub>(2),CH<sub>3</sub>CH=CH,-(3) C<sub>6</sub>H<sub>5</sub>CH=CH<sub>2</sub>.(4),CH<sub>3</sub>-CH<sub>2</sub>-CH<sub>2</sub>-(5), C<sub>6</sub>H<sub>5</sub>CH-(6)

**Table- 4.1:** Analytical Data and Other Physical Properties of Compounds (1-6)

No.	Compounds	%Yield	Colour	Melting point °C	% M		Molar conductance ohm <sup>-1</sup> cm <sup>2</sup> mol. <sup>-1</sup>
					Calculated	Found	
1	[Zn(C <sub>8</sub> H <sub>16</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	80	Light yellow	210	11.77	11.70	11.80
2	[Zn (C <sub>10</sub> H <sub>20</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	60	Red	200	11.20	111.22	12.08
3	[Zn (C <sub>14</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	75	Yellow	190	10.22	10.20	10.03
4	[Zn ( C <sub>24</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	70	White	215	10.28	10.25	12.03
5	[Zn (C <sub>20</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	85	Yellow	205	9.50	9.45	13.90
6	[Zn (C <sub>14</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	75	Light yellow	203	9.44	9.50	12.31

**Table- 4.2:** Elemental analysis data of compounds (1-6)

No.	Compounds	%C		%H		%N	
		Calculated	Found	Calculated	Found	Calculated	Found
1	[Zn(C <sub>8</sub> H <sub>16</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	18.39	18.35	2.06	2.05	21.45	21.00
2	[Zn (C <sub>10</sub> H <sub>20</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	20.68	20.60	1.72	1.72	19.31	19.03
3	[Zn (C <sub>14</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	26.41	26.40	2.20	2.21	17.61	17.20
4	[Zn ( C <sub>24</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	26.58	26.50	2.22	2.20	17.72	17.70
5	[Zn (C <sub>20</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	42.10	42.00	3.51	3.50	16.37	16.30
6	[Zn (C <sub>14</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	34.88	34.80	3.98	4.00	16.27	16.20

**Table- 4.3:** Magnetic moment data of compounds (1-6)

No.	Compounds	Sample length, l in cm	Weight of the sample, m in gm	Susceptibility of the empty tube, Ro	Susceptibility of the sample with tube ,R	Mass Susceptibility x <sub>2</sub> × 10 <sup>-4</sup> C.G.S.unit	Molecular weight,M	Molar Susceptibility x <sub>2</sub> × 10 <sup>-4</sup> C.G.S.unit	μ <sub>eff</sub> B.M
1	[Zn(C <sub>8</sub> H <sub>16</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	2.2	0.0695	-48	-22	1.71	552	0.943	1.50
2	[Zn (C <sub>10</sub> H <sub>20</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	2.1	0.0692	-47	-23	1.51	580	0.875	1.44
3	[Zn (C <sub>14</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	2.2	0.0596	-40	-21	1.46	636	0.928	1.49
4	[Zn ( C <sub>24</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	1.8	0.0559	-46	-24	1.47	632	0.929	1.49
5	[Zn (C <sub>20</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	1.8	0.0630	-42	-20	1.31	684	0.896	1.46
6	[Zn (C <sub>14</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	1.7	0.0589	-46	-23	1.38	688	0.949	1.50

**Table- 4.4:** Important infrared spectral bands of compounds (1-6)

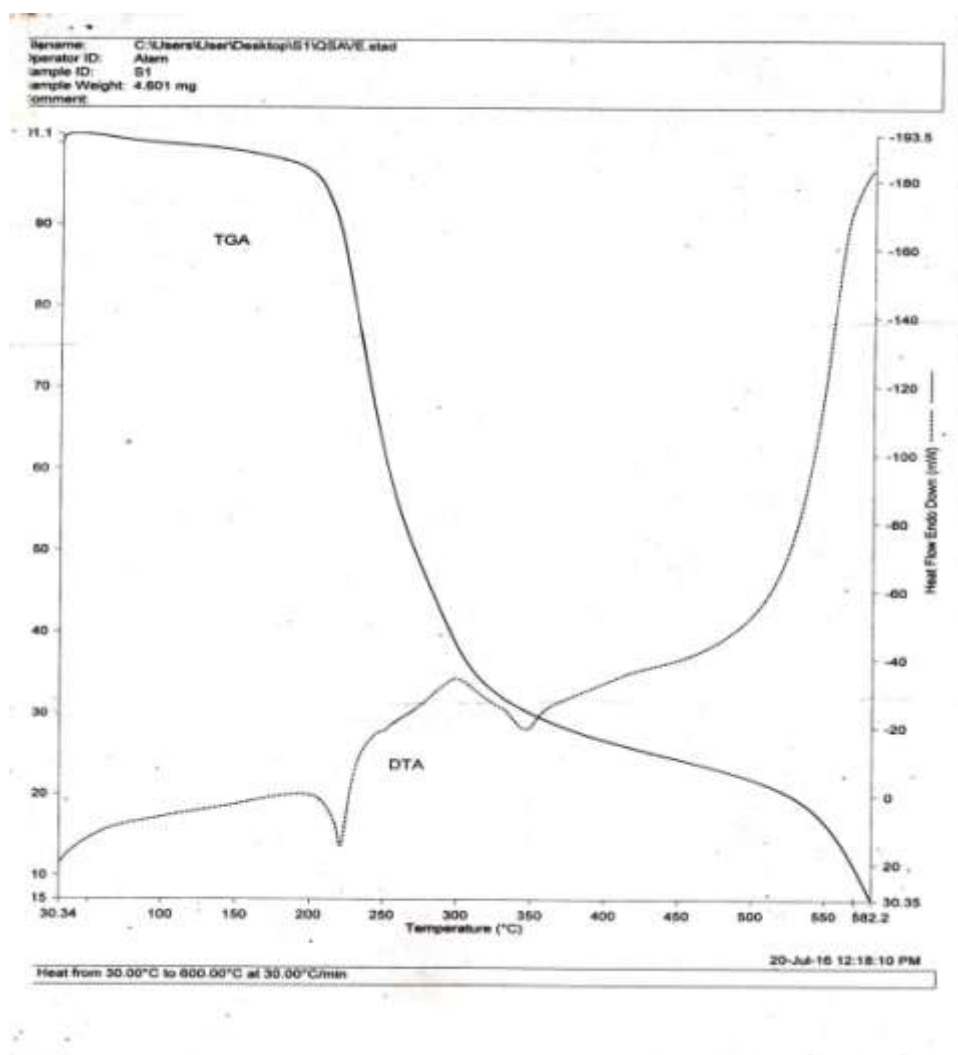
No.	Compounds	ν(C-H) cm <sup>-1</sup>	ν(C=O) cm <sup>-1</sup>	ν(N-H) cm <sup>-1</sup>	ν(M-N) cm <sup>-1</sup>	ν(ClO <sub>4</sub> ) cm <sup>-1</sup>
1	[Zn(C <sub>8</sub> H <sub>16</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	3047	1664	3251	420	1180,1089,623
2	[Zn (C <sub>10</sub> H <sub>20</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	2920	1649	3282	430	1105,979,623
3	[Zn (C <sub>14</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	2960	1640	3250	416	1150,1060,620
4	[Zn ( C <sub>24</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	2967	1650	3261	408	1160,1040,623
5	[Zn (C <sub>20</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	3040	1660	3254	409	1140,1080,621
6	[Zn (C <sub>14</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	3070	1680	3270	406	1170,1090,625

**Table- 4.5:** U.V- Visible Adsorption Maxima Of Compounds (1-6)

No.	Compounds	λ max (n,m)
1	[Zn(C <sub>8</sub> H <sub>16</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	310
2	[Zn (C <sub>10</sub> H <sub>20</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	310
3	[Zn (C <sub>14</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	300
4	[Zn ( C <sub>24</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	305
5	[Zn (C <sub>20</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	300
6	[Zn (C <sub>14</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	390

**Table- 4.6:** Thermal Analysis Data of Compounds (1-6)

No.	Compounds	% Ligand			%M Metal oxide(MO)		
		Tem <sup>0</sup> C	Calculated	Found	Tem <sup>0</sup> C	Calculated	Found
1	[Zn(C <sub>8</sub> H <sub>16</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	135.02	83.22	83.20	599.00	37.66	37.60
2	[Zn (C <sub>10</sub> H <sub>20</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	135.00	86.21	86.20	599.00	36.14	36.10
3	[Zn (C <sub>14</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	130.90	87.14	87.10	550.60	33.72	33.60
4	[Zn ( C <sub>24</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	130.45	88.91	88.50	450.00	29.06	29.00
5	[Zn (C <sub>20</sub> H <sub>28</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	140.50	88.23	88.20	530.00	30.86	30.40
6	[Zn (C <sub>14</sub> H <sub>24</sub> N <sub>8</sub> O <sub>4</sub> )(ClO <sub>4</sub> ) <sub>2</sub> ]	137.45	87.07	87.00	540.00	33.89	33.30



**Fig: 4.2-TGA, DTA**

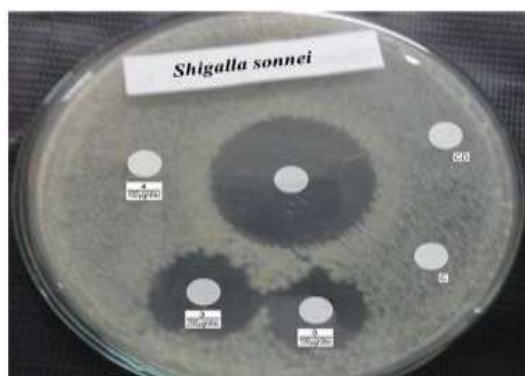
### Antibacterial Activity Testing

It has been observed that some drug increases the activity when administered as metal complexes or their metal chalets. The antibacterial activity of the metal complexes **1,2, 3, 4,5** and other complexes are recorded against fourteen pathogenic bacteria viz. *Salmonella-17*, *Klebsilla*, *Shigella dysenteriae*, *Shigella shiga*, *Shigella boydii*, *Shigella sonnei*, *Shigella flexneri*, *Escherichia coli*, *Pseudomonas aeruginosa*, *Salmonella*, *Bacillus megaterium*, *Sarcina lutea*, *Staphylococcus aureus*, *Bacillus cereus* And the result is given in (Table 5.1-5.2) the complex **1, 4** showed the most activities above fourteen pathogenic bacteria as shown (Fig 5.0). It is evident from all the tables that the under investigation showed the most activity compared to the complex **1, 2, 3, 4,5**

The malanodihydrized complexes **1, 3, 4** and **5** have shown good activity against the above fourteen pathogenic bacteria as seen in (Table 5.1-5.2). The complex **1** showed the best activity against, *Shigella boydii*, *Escherichia*, *Sarcina lutea coli* and less activity against *Shigella dysenteriae*. The complex **2** showed the best activity, *Pseudomonas aeruginosa* and less activity against *Salmonella-17* . The complex **3** showed the best activity, *shigella sonnei*, *Shigella shiga* and less activity against *Shigella dysenteriae*, *Escherichia coli*. The complex **4** showed the best activity against *Shigella boydii* and less activity against *Shigella flexneri*. The complex **5** showed the best activity against *Bacillus megaterium* and less activity against *Sarcina lutea* .The complex **6** are not showed good activities against the above fourteen pathogenic bacteria. The complex **1** showed good activities *Shigella boydii* and less activity against *Shigella dysenteriae*. The complex **2** showed good activities *Pseudomonas aeruginosa* and less activities against *Salmonella-17*. The complex **3** showed good activitie *Shigella shiga* and less activities against *Klebsilla* . All the result are compared with the standard compound, kanamycin as seen in the Table (5.1-5.2) the ligand malanodihyrazide ( $C_3H_8N_4O_2$ ) did not show any activities against the above fourteen pathogenic bacteria.

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From here it is concluded that the complex **1, 3** and **4** showed good activities against the fourteen pathogenic bacteria as compared to the standard compound, kanamycin. It is evident that the ligand malanodihydrazide did not show any activity.



**Fig: 5.0:** Photographic representation of zone of inhibition of the complexes 3 the standard compound kanamycin against Shigalla sonnei

**Table-5.1**

Name of microorganisms Name of test sample	Diameter of zone of inhibition (mm)									
	Complex01			Complex02			Complex03			Stand 30
Done	200µg /disc	100µg /disc	50 µg /disc	200µg /disc	100µg /disc	50 µg /disc	200µg /disc	100µg /disc	50 µg /disc	KAN
<b>Gram negative bacteria</b>										
1. Salmonella-17	15	9	3	12	6	2	17	8	4	20
2. Klebsilla	16	7	3	10	5	2	8	2	-	20
3. Shigella dysenteriae	12	5	2	14	6	3	8	5	2	20
4. Shigella shiga	16	7	4	13	6	3	18	12	4	20
5. Shigella boydii	18	10	4	10	5	2	10	4	2	20
6. Shigella sonnei	16	7	3	15	7	3	18	14	4	20
7. Shigella flexneri	15	6	2	14	7	2	7	3	2	
8. Escherichia coli	18	10	5	16	7	3	8	4	2	20
9. Pseudomonas aeruginosa	16	8	4	18	8	3	16	8	4	20
10. Salmonella	15	7	3	12	6	3	10	5	2	20
<b>Gram positive bacteria</b>										
11. Bacillus megaterium	16	10	4	15	7	3	15	6	3	20
12. Sarcina lutea	18	12	5	13	6	2	10	6	2	20
13. Staphylococcus aureus	15	6	3	14	6	3	17	9	6	20
14. Bacillus cereus	16	8	4	12	6	2	10	5	2	20

**Table-5.2**

Name of microorganisms Name of test sample	Diameter of zone of inhibition (mm)									
	Complex04			Complex05			Complex06			Stand 30
Done	200µg /disc	100µg /disc	50µg /disc	200µg /disc	100µg /disc	50µg /disc	200µg /disc	100µg /disc	50µg /disc	KAN
<b>Gram negative bacteria</b>										
1. Salmonella-17	16	8	3	15	6	2	-	-	-	20
2. Klebsilla	10	4	2	16	8	3	-	-	-	20
3. Shigella dysenteriae	15	7	2	14	7	2	-	-	-	20
4. Shigella shiga	16	7	3	14	7	2	-	-	-	20
5. Shigella boydii	18	9	4	16	8	3	8	3	-	20
6. Shigella sonnei	12	5	2	15	7	3	-	-	-	20
7. Shigella flexneri	10	4	2	12	6	2	8	3	-	20
8. Escherichia coli	15	4	2	14	6	3	-	-	-	20
9. Pseudomonas aeruginosa	15	9	3	15	7	3	-	-	-	20
10. Salmonella	12	8	4	18	8	3	-	-	-	20
<b>Gram positive bacteria</b>										
11. Bacillus megaterium	12	6	3	16	7	3	8	-	-	20
12. Sarcina lutea	14	7	2	12	5	2	-	-	-	20
13. Staphylococcus aureus	13	6	2	14	7	3	-	-	-	20
14. Bacillus cereus	10	4	2	16	8	3	-	-	-	20

The present work also determined the minimum inhibitory concentration of the more active complexes 1, 2, 3, 4, 5 by a serial dilution method. The tube of broth medium (1mL) containing graded doses of sample were incubated with the test organisms. After suitable incubation growth occurred in these inhibitory tubes, where the concentration of the sample was below the inhibitory level, the culture became turbid (cloudy). The growth of the microorganisms was not observed above the inhibitory level and the growth of the microorganisms was not observed above the inhibitory level and the tubes remained clear. The minimum inhibitory results are furnished in Table-5.3.

**Table-5.3**

Test organism	Complex 1	Complex 2	Complex 3	Complex 4	Complex 5
	MIC( $\mu$ g/mL)				
<i>Salmonella-17</i>	32	32	64	32	32
<i>Klebsilla</i>	32	32	64	32	32
<i>Shigella dysenteriae</i>	16	32	64	32	32
<i>Shigella shiga</i>	32	32	64	32	32
<i>Shigella boydii</i>	32	32	64	32	32
<i>Shigella sonnei</i>	32	16	64	16	32
<i>Sigella flexneri</i>	32	32	64	32	32
<i>Escherichia coli</i>	32	32	64	32	16
<i>Pseudomonas aeruginosa</i>	32	32	64	32	32
<i>Salmonella</i>	32	32	64	32	32
<i>Bacillus megaterium</i>	64	32	64	32	32
<i>Sarcina lutea</i>	32	64	64	64	64
<i>Staphylococcus aureus</i>	32	32	32	32	32
<i>Bacillus cereus</i>	32	32	64	32	32

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