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X-RAYDIFFRACTION STUDIES OF Cu(II), Zn(II), Mo(II), Fe(II) COMPLEXES WITHGLIBENCLAMIDE(5-CHLORO-N-(4-[N-(CYCLOHEXYLCARBONYL)SULFAMOYL]PHENETHYL)- 2-METHOXYBENZAMIDE,AN ORAL ANTIDIABETIC DRUG

Mohammad Tawkir Sheikh and Sushil B. Kapoor
Department of Chemistry, Arts,Comm.and Sci.College,Tukum,Chandrapur-442401 (India)

Abstract
Glibenclamide (5-Chloro-N-(4-{N-(Cyclohexyl Carbonyl) Sulfamoyl]Phenethyl)-2-methoxybenzamide was used to synthesize Cu(II), Zn(II), Mo(II), Fe(II) complexes. Metal complxes were characterized by elemental analysis, IR, TGA. The crystal structure of complexes were further determined by X-ray diffraction method. The XRD data was used to calculate various parameters like crystal system, volume,density,porosity,particlesizeetc.which shows that the complexes of Cu(II) and Zn(II) are **tetragonal** while the complexes ofMo(II)and Fe(II) shows **octahedral structure**.
Key words: Glibenclamide, Crystal structure, Cu(II), Zn(II), Mo(II), Fe(II) complex.

Introduction

Polyfunctionallyrings compounds and synthesis of their metal complex which have various biological activities and include hetero atom, have been formed in organic synthesis and coordination chemistry.[1-6]Manytrasition and inner trasition metal complexes have been synthesized for analytical and commercial applications many of medicinal use.[7-9] literature survey reveals that the transition metal complexes generally crystallized with tetrahedral, octahedral geometry.[10-12]

Experimental

All the chemicals used for the preparation of complexes are of Hi-media AR grade E-merk. Metal complexes are synthesized by adding metal salt solution in appropriate solvent to the solution of the ligand. The mixture was refluxed for 3-4 hours. Then the precipitate of metal complxes was obtained. It is filtered, washed and dried in vacuum desiccators.
All selected metals forms 1:2 complexes with glibenclamide were confirmed by to be method as modified by Turner and Anderson.[13-14]

Results and Discussion:

The result of ESR spectra and X-ray diffraction of Cu(II), Zn(II), Mo(II), Fe(II) complexes with Glibenclamide were obtained and summarized in following tables. Al reflections has been indexed for ***h, k, l*** values using reported literature[15-23]and full proof suit XRD software v.2.0 by using foolproof suite XRD software the d-values of metal complexes were obtained. From ESR spectra of complexes the value of g_1,g_2,g_3 and g_{av} can be determined. This value are tabulated in table No. (2)
In case of Cu(II) complexg_{av} value is 2.23 which is less than 2.271. the values of indicates the presence of sufficient covalence between the metal ion and donar atom.[24-25]
In case of Zn(II) complex gav value is found to be 2.21. this value is less than 2.25. it is assignable to the presence of covalent character in metal ion and donar atom. In case of Mo(II) complex and Fe(II) complex g_{av} values found to be 2.30 and 2.09 respectively. This valuesless than 2.387 and 2.247 indicates presence of covalent characters in coordinate bond.

3.1 X-ray diffraction studyof Glibenclamide complexes

The X-ray diffraction pattern of Cu(II), Zn(II), Mo(II) and Fe(II) complexes has been determined 2θ range from 5.0084 to 79.97884°,Diffractograms (Fig-1,2,3,4) and data has been summarized in the followingtable.No.3,4,5and6.

Table 1: Physico-chemical and Analytical data of GlibenclamideComplexes.

Sr. No.	Composition of complex	Metal Ligand Ratio	Colour	% Yield	M.P. (°C)	% of Metal observed/ Required
01	(C23H27O5ClN3S)2Cu	1:2	Green	63	188	4.041(5.50)
02	(C23H27O5ClN3S)2Zn	1:2	White	55	205	4.698(5.84)
03	(C23H27O5ClN3S)2Mo2H2O	1:2	Green	52	185	7.025(8.11)
04	(C23H27O5ClN3S)2Fe2H2O	1:2	Brown	49	189	8.613(8.64)

Sr. No.	% of Carbon observed/ (Required)	% of H observed/ (Required)	% N observed/ Required	% of S observed/ Required	Stability constant log k lit/mole	Free Energy Change (-ΔF)
01	46.5 (49.70)	4.50 (4.86)	7.60 (7.57)	5.11 (5.77)	10.7219	-14.7438
02	43.72 (49.70)	4.99 (4.86)	7.34 (7.75)	5.90 (5.80)	10.6973	-14.7099
03	46.79 (49.70)	4.082 (4.86)	6.25 (7.70)	5.81 (5.81)	10.6677	-14.6692
04	48.01 (49.70)	4.616 (4.86)	6.56 (7.78)	5.88 (5.81)	10.69618	-14.70389

Table 2: ESR data of Cu(II), Zn(II), Mo(II) and Fe(II) complexes.

ESR spectral parameters	$[(GLB)_2Cu]$	$[(GLB)_2Zn]$	$[(GLB)_2 Mo_2H_2O]$	$[(GLB)_2Fe_2H_2O]$
g_1	1.807	1.700	1.910	1.756
g_2	1.960	1.848	2.066	1.919
g_3	2.11	2.001	2.228	2.081
gav	2.271	2.252	2.387	2.247
	2.23	2.21	2.30	2.09

Table 3: Cell data and crystal parameters for $[(GLB)_2Cu]$ complex

a(Å) = 21.6891
b(Å) = 23.1891
c(Å) = 27.6065
Standard deviation = 0.0026%
α=90°, β=90°, λ=90°

Volume (abc)Å = 13884.253
Dcal = 7.4434 g/cm^3
Dobs =7.39890 g/cm^3
Crystal system = Orthorombic(Tetragonal)
Porosity(%) = 8.190
Density = 0.085366g/cm^3
Particle size = 9.3780 microns
Space group = Pbnm,

2 θ	I/I$_0$	D(Obs)	D(Cal)	h	k	l
11.1140	12.32	7.96124	7.95674	1	1	3
11.9617	100.00	7.39890	7.44341	0	3	1
12.4745	38.63	7.09587	7.04036	1	3	1
16.4931	79.28	5.37488	5.37099	0	1	5
19.2866	96.20	4.60223	4.60093	0	0	6
19.7025	29.37	4.50601	4.50078	1	0	6
21.2609	85.27	4.17912	4.1667	2	1	6
23.1534	20.67	3.84163	3.84938	0	5	4
23.4613	29.67	3.79190	3.79015	1	5	4
24.8712	17.52	3.58006	3.58425	0	1	2
28.0146	12.36	3.18510	3.18605	3	2	2
30.6354	19.22	2.91832	2.91801	0	7	2
31.998,	59.96	2.97708	2.79437	4	7	2

From the cell data and crystal lactice one can conclude thatCu(II) complex is having ***Orthorombic crystal*** system

Table4: Cell data and crystal parameter for $[(GLB)_2Zn]$ complex

a(Å) = 21.6891
b(Å) = 23.1891
c(Å) = 27.6065
Standard deviation = 0.00276%
α =90°, β=90°, γ=90°

Volume (abc)Å = 13884.253
Dcal = 3.21627 g/cm^3
Dobs =3.13908 g/cm^3
Crystal system =Orthorhombic(Tetragonal)
Porosity(%) = 8.1371%
Density = 0.08847g/cm^3
Particle size =8.1371 microns
Space group = Pbnm,

2 θ	**I/I$_0$**	**D(Obs)**	**D(Cal)**	**h**	**k**	**l**
8.3049	17.35	10.64676	10.68994	-2	1	2
10.8705	100.00	8.13908	8.21627	1	2	2
12.3720	49.65	7.15443	7.20777	0	2	3
14.1502	11.73	6.25913	6.29442	2	-3	0
14.6554	21.13	6.04446	6.00282	-2	-2	3
16.1775	45.61	5.47904	5.18884	1	-4	1
16.3942	73.98	5.40710	5.42228	4	0	0
17.1632	24.84	5.16651	5.14805	0	3	4
18.7322	37.43	4.73716	4.78419	-1	4	3
19.9779	21.44	4.44450	4.43900	4	3	0
20.4316	17.67	4.34686	4.34882	1	4	4
21.5818	35.26	4.11770	4.10814	2	-4	4
22.0073	25.85	4.03909	4.07423	2	5	2
23.2671	16.03	3.82312	3.82684	1	1	7
24.3024	16.13	3.66254	3.66810	1	0	2
25.5043	27.41	3.49261	3.49608	1	2	1
28.3278	16.14	3.15059	3.15097	0	3	8
32.1148	63.24	2.78719	2.78063	2	8	1
32.7844	52.50	2.57916	2.57895	3	0	10
36.6116	83.79	2.45452	2.45585	8	4	0
56.9255	24.44	1.61762	1.61773	4	2	2
63.0934	16.06	1.47352	1.4289	4	4	0
68.2744	17.27	1.35100	1.3662	5	3	1

From above data it is clear that Zn(II) complex is having ***Orthorhombic crystal*** *system*.

Table 5: Cell data and crystal parameters for [(GLB)$_2$Mo2H$_2$O] complex

a(Å) = 21.7628	Volume (abcsinβ)Å	= 14071.30322
b(Å) = 23.4281	Dcal	= 13.80660 g/cm^3
c(Å) = 27.6010	Dobs	= 13.85955 g/cm^3
Standard deviation = 0.0034%	Crystal system	= Monoclinic(Octahedral)
α =90°, β=89.2°, γ =90°	Porosity(%)	= 3.8795
	Density	= 0.086648g/cm^3
	Particle size (t)	=23.5720
	Space group	= Pmmm,

2 θ	**I/I$_0$**	**D(Obs)**	**D(Cal)**	**h**	**k**	**l**
6.3774	100.00	13.85955	13.80660	1	1	1
10.8970	26.15	8.11932	8.26023	1	-2	2
11.4807	3.82	7.70780	7.65932	2	-2	1
14.4063	5.14	6.14843	6.16741	3	2	0
15.2123	5.68	5.82442	5.82736	2	0	4
16.9963	15.53	5.21656	5.21643	1	1	5
19.2834	30.43	4.60299	4.60017	0	0	6
19.9557	28.96	4.44940	4.43686	0	5	2
22.2511	26.35	3.99534	3.98622	4	4	0
23.4571	3.85	3.79257	3.80192	5	3	0
24.7115	3.22	3.60283	3.59621	6	0	1
28.6699	4.14	3.11376	3.11288	1	7	3
32.1833	1.47	2.78142	2.78123	3	7	4
33.5511	3.97	2.67108	2.67159	4	0	9
35.2684	2.14	2.54486	2.54429	3	4	9

From the cell data and crystal lactice one can conclude that Mo(II) complex is having ***Monoclinic crystal*** system.

Table 6: Cell data and crystal parameters for [$(GLB)_2Fe2H_2O$] complex

a(Å) = 21.7621	Volume (abcsinβ)Å	= 14065.269
b(Å) = 23.4271	Dcal	= 2.77740 g/cm^3
c(Å) = 27.5913	Dobs	= 2.77239 g/cm^3
Standard deviation = 0.0034%	Crystal system	= Monoclinic(Octahedral)
α =90°, β=89.2°, γ =90°	Porosity(%)	= 3.78 %
	Density	= 0.086648g/cm^3
Space group = Pm	Particle size	=23.5720 microns

2 θ	**I/I$_0$**	**D(Obs)**	**D(Cal)**	**h**	**k**	**l**
19.1026	84.70	4.64230	4.65280	-2	3	4
23.2221	14.29	3.82726	3.82021	1	5	4
28.0902	44.86	3.17406	3.17088	5	5	1
29.0522	38.56	3.07111	3.07309	6	4	1
32.2038	100.00	2.77739	2.77740	0	5	8
33.9247	47.03	2.64033	4.64037	4	7	4
38.7004	25.14	2.32479	2.32319	7	0	8
48.8725	33.07	1.86206	1.86187	7	0	8
54.6565	13.48	1.67789	1.67787	3	1	0
59.5637	12.38	1.55084	1.5508	0	1	2

From the cell data and crystal lattice one can conclude that Fe(II) complex is having ***monoclinic crystal*** system.

Table (7)

Complexes	**Mol^r Formulae**	**Mol^r Weight (gm/mole)**	**Crystal/ System**
($C_{23}H_{28}O_5ClN_3S$**)**$_2Cu$	$C_{46}H_{56}O_{10}Cl_2N_6S_2Cu$	1051.548	Orthorhombic
($C_{23}H_{28}O_5ClN_3S$**)**$_2Zn$	$C_{46}H_{56}O_{10}Cl_2N_6S_2Zn$	1051.378	Orthorhombic
($C_{23}H_{28}O_5ClN_3S$**)**$_2Mo2H_2O$	$C_{46}H_{60}O_{12}Cl_2N_6S_2Mo$	1083.954	Monoclinic
($C_{23}H_{28}O_5ClN_3S$**)**$_2Fe2H_2O$	$C_{46}H_{60}O_{12}Cl_2N_6S_2Fe$	1079.853	Monoclinic

Figure -1

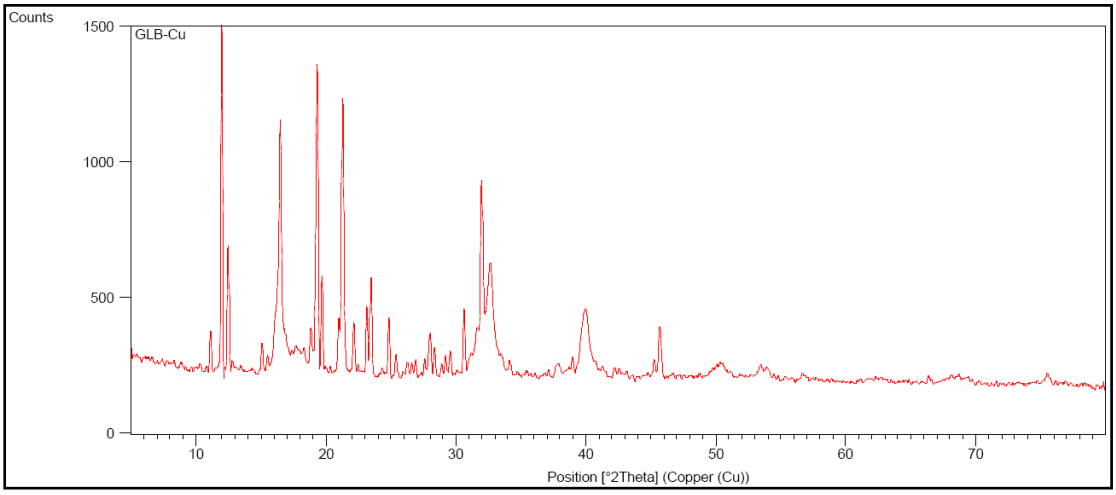


Figure -2

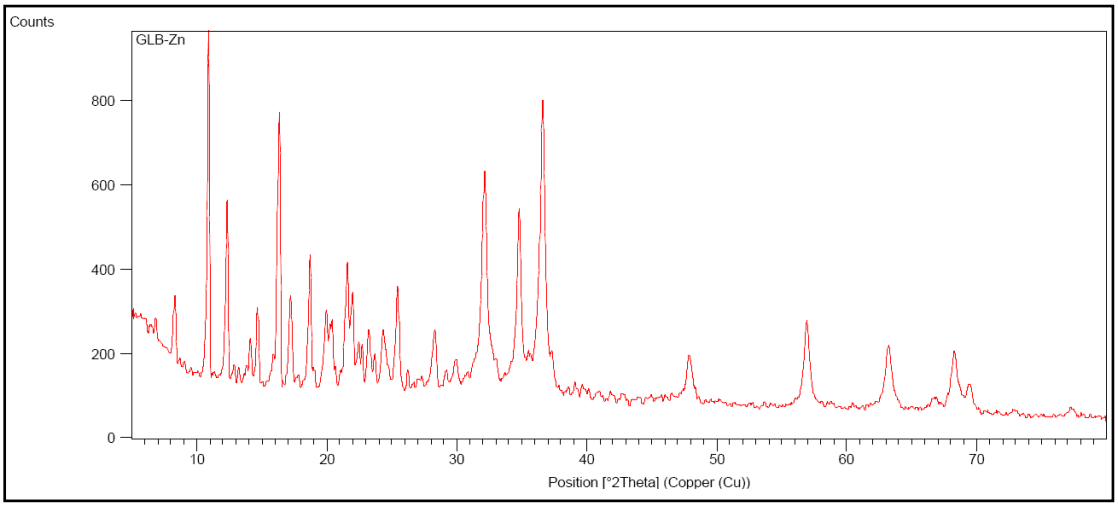


Figure -3

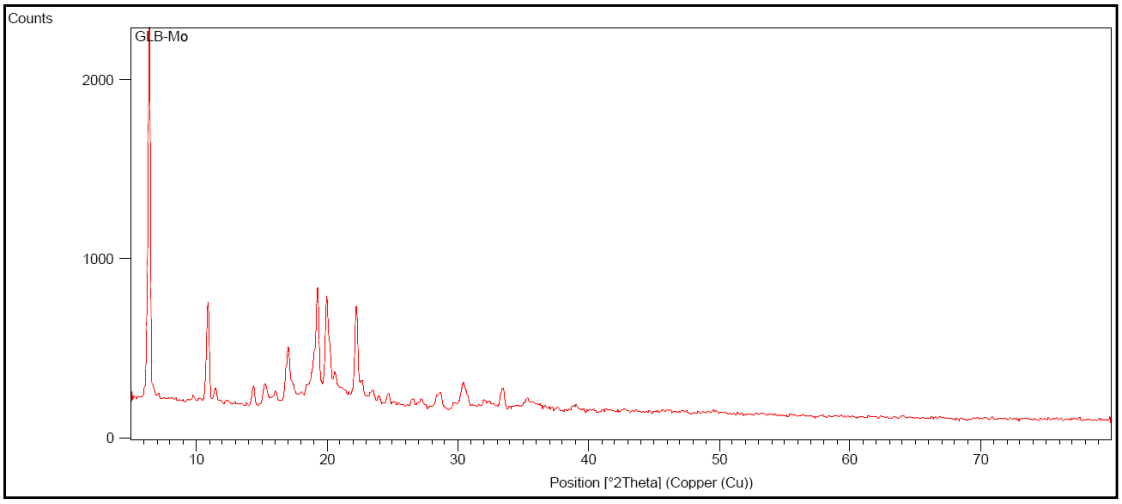
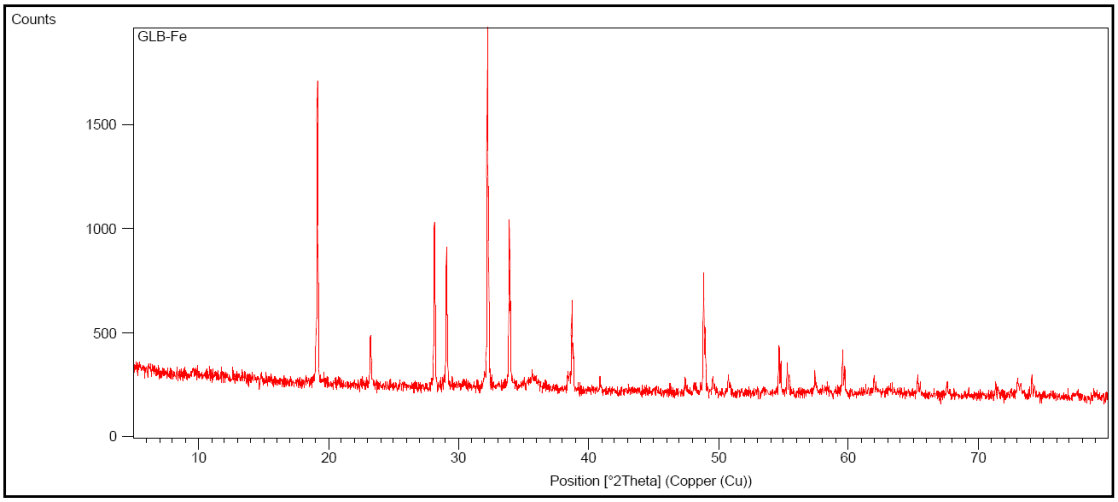
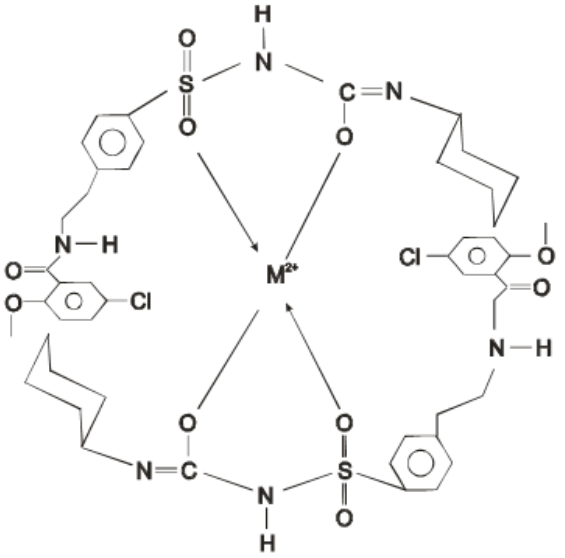


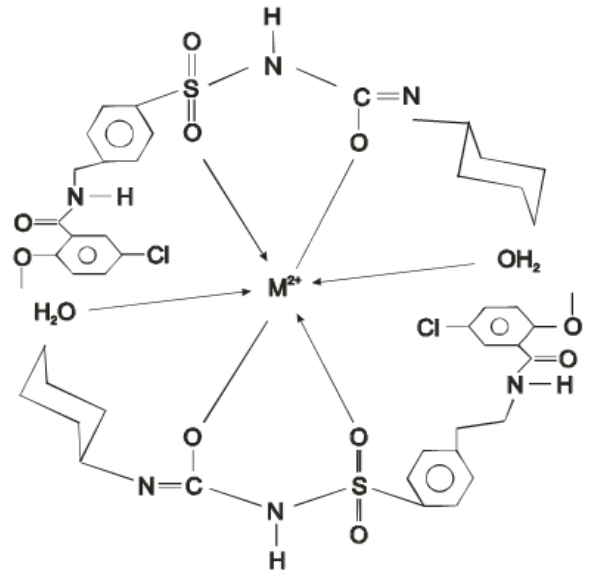
Figure -4





Where, M = Cu, Zn

Figure -5 Proposed structure of $(GLB)_2Cu$ and $(GLB)_2Zn$ complexes



Where, M = Mo and Fe

Figure -6 Proposed structure of $(GBM)_2Fe2H_2O$ and $(GBM)_2Mo2H_2O$ complexes

Conclusion

X-ray diffraction studies also confirms the complexes and formation of new bonds. The number of peaks in Glibenclamide are 11 ively while that of $(GLB)_2Cu$, $(GLB)_2Z_n$ $(GLB)_2Mo.2H_2O$ and $(GLB)_2Fe.2H_2O$ are 13,23,15 and 10 respectively (Fig-5) and number of peaks in case of copper chloride and zinc acetate are 8 and 7 respectively. Thus indicating that complexes formed are a well kit one moreover the X-ray pattern of neither Glibenclamide nor copper chloride and zinc acetate are seen in diffractogram of complexes. all the reflections present are new ones and the patterns are fairly strong. On comparing the pattern obtained with available literature. It is evident that its pattern is not in good agreement with available information and thus confirms the formation of totally new complexes The X-ray pattern have been indexed by using computer software(FPSUIT 2.0V) and applying interactive trial and error method keeping in mind the characterstics of the various symmetry system,till a good fit was obtained between the observed and the calculated $Sin^2\theta$ value.The unit cell parameters were calculated from the indexed data, from cell data and crystal lattice parameters of $(GLB)_2Cu$, $(GLB)_2Z_n$) indicates complexes attributed to Orthorhombic crystal system.While$(GLB)_2Mo.2H_2O$ and $(GLB)_2Fe.2H_2O$ complexes attributed to Monoclinic crystal system Table-7..

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