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Synthesis, characterization and antimicrobial activity of new homo-or hetrotrinuclear Cu(II) and Mn(II) biazodianil complexes

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ABSTRACT

A Series of homo or hetro di and trimetalic complexes has been prepared containing both Cu(II) and Mn(II) ions with bisazodianil ligands based on 2 carboxy, 3′carbonyl,4′. 6′dihydroxy azo benzene ($H_3 L^1$) or 2 hydroxy, 3′carbonyl ,4′,6′dihydroxy azo benzene($H_3 L^3$). The complexes prepared were characterized by elemental anlysis TGA, DTA, IR, UV/ Vis and ESR spectra as well as magnetic moment and atomic absorption determinations. Antimicrobial activity of the ligands and their complexes have been examined against the *bacterial strains Bacillus subtilis*, *E.coli*, *Staphylloccus aerues*, *Pesudomonas auregonosa* and *Salmonella* spp. The results show that the complexes are potent antimicrobial than the parent ligand .These complexes have been found to exhibit moderate to strong antimicrobial activity against the tested microbes.

INTORDUCTION

The term “dinucleating ligands” was first introduced in 1970 by Rebson[1] to describe the class of polydentate chelating agents, able to bind simultaneously two metal ions. Since then, a very large unmber of such ligands were designed, and their coordination compounds were thoroughly investigated. Many workers have reported homo and[2-5] hetero dimetallic complexes. Dinuclear metal complexes are important in the investigation of the interaction between metal ions in dinclear metal center of proteins[6,7]. The possible applications of the complexes with this type of ligands vary from modeling the active sites of many metalloenzymes[8,9] to hosting and carrying small molecules[10-11] during catalysis[12]. Three new Co(II) and Mn (II) complexes with the phenol-based on ligand Hpyzald which contains the formyl group were reported[13,14]. The preparation and study of inorganic compounds containing biologically important ligands is made easy because certain metal ions are active in many biological processes. The fact that copper with other inororganic metallic elements are essential and exhibit great biological activity when associted with certain metal complexes, participating in oxygen transport, electronic transfer reactions or the storage ions [15]. The bis-azodanils metal complexes have been subject of some recent interstigations due to their excellent catalytic properties and biological activity. This has prompted much increasing effort in the development of new bisazodianil complexes. Our recent focus has been located on homo or hetero bimetallic bisazo-dianil complexes possessing simple metal ions[16]. The aim of this study is the synthesis, characterization and biological evalution of mono, dicopper (II) and hetrotrinuclear Cu (II) and Mn (II) biazodianil complexes. The prepared complexes showed a good biological activity.

EXPERIMENTAL

Preparation of organic ligands

The azo dyes based on anthranilic acid or o-aminophenol and 2,4 dihydroxbenzaldhyde were as given previously[17].

Prepanation of complexes

1. Preparation of mono Cu (II) complexes

10^{-3} mole of $CuCl_2$.2 H_2O was dissolves in the least amount of hot ethanol, then it was mixed with a similar solution containing 10^{-3} mole of 2 carboxy,3′carbonyl, 4′.6′dihydroxy azo benzene (H_3 L^1) or 2 hydroxy, 3′carbonyl ,4′,6′dihydroxy azo benzene (H_3 L^3) .The reaction mixture was refluxed in a water bath for 6 hr. On cooling the Cu(II) complex then seperated, which was filtered off and dried (A for H_3 L^1 and D for H_3 L^3).

2. Preparation of di Cu(II) complexes

2×10^{-3} of mono copper complex (A or D) was dissolved in the least amount of hot ethanol, then the solution was mixed with 10^{-3} mole of propenyl diamine and the mixture was reflexed for 6 hrs. in a water bath. On cooling the mixture, the binuclear Cu (II) complex separated which was filtered off, dried (B and E).

3. Preparation of di copper and Mn (II) complexes

10^{-3} mole of $MnCl_2.4H_2O$ was dissolved in the least amount of hot ethanol. Then it was mixed with a similar solution containing 10^{-3} mole of the dicopper complex (B or E).The mixture was refluxed in a water bath for 6 hrs.on cooling the solution, the trinuclear complexes (C and F) separated which were filtered off and dried.

The apparatus and working procedures for the characterization of the complexes were the same as previously described[18].

Preparation of microbial cultures

Bacillus subtilis, *E.coli*, *Staphylloccus aerues*, *Pesudomonas auregonosa* and *Salmonella* spp. were used as the test organisms.The bacterial strains were inoculated into nutrient broth and incubated for 24 hours. In the well diffusion method,the sterile agar (oxoid) for bacteria were inoculated with the test microogranisms. The compounds dissolved in DMF as 0.1% and the sterial solution were added in wells (4mm diameter) in agar plate media, and the plates were incubated at 35°C for 24 hours.The resulting inhibation zones on the plates were measured after 2 days. The data reported in TABLE 6 are the average data of three experiments.

RESULTS AND DISCASSION

The data of elemental analysis (TABLE 1) support the suggested formululas for the complexes. The calculated and found values are in a good agreement with each other.

Thermal analysis

1. Thermogravimetic analysis

Thermogravimetric analysis (TGA) yielded the results showen in TABLE 2. For all investigated complexes, the thermograms exhibited the following commn features:

a. The lattic water or ethanol molecules in the complexes were evaporated with in the temperature range 50-125°C, while coordinated water molecule

TABLE 1: Elemental analysis, magnetic moment , ESR, M.p. °C and yield% data

Complex	C%	H%	Cu%	Mn%	H_2O%	EtOH%	g_{eff} Cu	g_{eff} Mn	μ_{eff}	Yield%	m.p °C
A $[CuHL^1(H_2O)_3].2H_2OEtOH$	(39.02) 39.7	(3.89) 4.9	(13) 13.1		(18.6) 18.6	(8.88) 9.5	1.84		0.72	70	125
B $[Cu_2HL^2(H_2O)_6].3H_2O$	(41.7) 41	(4.1) 4.4	(13.04) 14		(17.4) 16.5		1.8		0.72	79	Over 260
C $[Cu_2 MnL^2(H_2O)_8].4H_2O$	(34.97) 34.5	(4.1) 4.2	(11.2) 11.7	(4.9) 5.1	(22.6) 22.5		1.82	1.62	2.65	60	Over 260
D $[CuHL^3(H_2O)_3]. H_2O$	(39.85) 39.84	(3.98) 4	(15.97) 16.2		(18.4) 17.6		1.82		0.73	72	120
E $[Cu_2HL^4(H_2O)_6].5H_2O$	(42.5) 42.2	(4.58) 4.6	(14) 13.79		(20.61) 20.6		1.82		0.77	60	Over 260
F $[Cu_2 MnL^4(H_2O)_8].8H_2O$	(33.1) 33.6	(4.26) 5.2	(11.2) 12.2	(5.6) 5.4	(28.1) 28.2		1.84	1.62	2.53	60	Over 260

(found) calculated

TABLE 2 : The TGA thermal data of complexes investigated

Complex	Step	$t_i{}^0C$	$t_m{}^0C$	$t_f{}^0C$	Mass loss %(found)	Assignment
A $[CuHL^1(H_2O)_3].2H_2OEtOH$	1 st	36	81.0	127	9 (8.8)	Loss of lattic ethanol molecules
	2 nd	127	145	163	7.4 (7.4)	Loss of lattic waterl molecules
	3 rd	163	181	199	11.1 (11.2)	Loss of coordinated water molecules
	4 th	199	510	821	-	Decomposition and formation of CuO
C $[Cu_2 MnL^2(H_2O)_8].4H_2O$	1 st	28	55.5	83	8.3 (8.2)	Loss of lattic water molecules
	2 nd	99	132.5	166	3.3 (3.5)	Loss of coordinated water molecules of Mn(II)
	3 rd	166	99	232	9.9 (10)	Loss of coordinated water molecule of two Cu(II)
	4 th	232	489.5	747	-	Decomposition and formation ofCuO and MnO
D $[CuHL^3(H_2O)_3]. H_2O$	1 st	20	37.5	55	4.4 5.3)	Loss of lattic water molecules
	2 nd	55	100	145	13.1(13. 2)	Loss of coordinated water molecules
	3 rd	145	345	545	-	Decomposition and formation of CuO
F $[Cu_2 MnL^4(H_2O)_8].8H_2O$	1 st	27	71.5	116	12.5(10.5)	Loss of lattic water molecules
	2 nd	116	141	166	3.4 (3)	Loss of coordinated water molecules of Mn(II)
	3 rd	182	207	232	10.4 (10.5)	Loss of coordinated water molecule of two Cu(II)
	4 th	232	373	514	-	Decomposition and formation ofCuO and MnO

TABLE 3 : Thermo-kinetic parameters of the complexes investigated from TGA

Complex	Step	n	r	E* KJ/mole	ΔH* KJ/mole	ΔS*J/mole	ΔG* J/mole
A $[CuHL^1(H_2O)_3].2H_2OEtOH$	1 st	1	0.9925	32.94	30.02	-40.9	44.5
	2 nd	0.66	0.89810	119.16	114.55	-42.2	132.1
	3 rd	0.5	0.9744	93.93	89.31	-42.9-	108.7
	4 th	0	0.8694	15.81	11.2	-47.8	50.3
C $[Cu_2 MnL^2(H_2O)_8].4H_2O$	1 st	1	0.9940	45.21	42.29	-40.2	55.5
	2 nd	0.66	0.9999	31	26.38	-42	43.42
	3 rd	1	0.9907	120.8	116.8	-43.3	36.6
	4 th	0.33	0.9858	7.83	3.21	-47.2	39.26
D $[CuHL^3(H_2O)_3]. H_2O$	1 st	1	0.9512	58.183	55.26	-39.8	67.5
	2 nd	0.66	0.9843	25.88	21.26	-41.3	36.4
	3 rd	0	0.9316	2.46	2.15	-45.5	30.26
F $[Cu_2 MnL^4(H_2O)_8].8H_2O$	1 st	1	0.9778	46.76	43.84	-40.6	57.8
	2 nd	0.66	0.9585	76.31	71.70	-42.2	89.1
	3 rd	0	0.9994	80.84	76.02	-43.4	96.8
	4 th	0	0.9907	17.35	12.7	-45.9	42.35

were removed at 150-250 °C.The number of water or ethanol molecules in the complexes was determined from mass loss on the thermogram The mass losses for the coordinated water molecules in case of complexes C and E reveals that water coordinated to the manganese ions is removed at lower temperature than that coordinated to the copper ions.This reflects stronger bonding of coordinated water to the copper ion than for the manganese ion.

b. The last step in the decomposition reaction took place at a temperature higher than 300°C involved the combustion of the organic ligand leading to the formation of the metal oxides as final decomposition product.The metal content was calculated from the weight of the final residue. The results were in satisfactory agreement with those of atomic absorption analysis.

The order, n, and activation energy, E*, of the decomposition steps for some selected complexes were determined from TGA results using the Coats-Redfern equation[19].

$$\ln [1-(1-\alpha)^{1-n}]/ (1-n)\, T^2 = M/T + B \quad \text{for } n \neq 1 \qquad (1)$$

$$\ln [-\ln (1-\alpha)]\, (1-n)\, T^2 = M/T + B \quad \text{for } n = 1 \qquad (2)$$

where $M = -E^*/R$ and $B = \text{In } AR/\varphi.E^*$; E^*,R, A and φ are the activation energy, gas constant, pre-exponential factor and heating rate respectively.

The correlation coefficient, r, is computed using the least squares method for different values of n. The left hand side of equations (1) and (2) were plotted versus 1/T for different n values (0-1), the n value which gave the best fit (r≈1) was chosen as the order parameter for the decomposition stage of interest. From the inter-

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	2 nd	0.66	0.89810	119.16	114.55	-42.2	132.1
	3 rd	0.5	0.9744	93.93	89.31	-42.9-	108.7
	4 th	0	0.8694	15.81	11.2	-47.8	50.3
C $[Cu_2 MnL^2(H_2O)_8].4H_2O$	1 st	1	0.9940	45.21	42.29	-40.2	55.5
	2 nd	0.66	0.9999	31	26.38	-42	43.42
	3 rd	1	0.9907	120.8	116.8	-43.3	36.6
	4 th	0.33	0.9858	7.83	3.21	-47.2	39.26
D $[CuHL^3(H_2O)_3].\ H_2O$	1 st	1	0.9512	58.183	55.26	-39.8	67.5
	2 nd	0.66	0.9843	25.88	21.26	-41.3	36.4
	3 rd	0	0.9316	2.46	2.15	-45.5	30.26
F $[Cu_2 MnL^4(H_2O)_8].8H_2O$	1 st	1	0.9778	46.76	43.84	-40.6	57.8
	2 nd	0.66	0.9585	76.31	71.70	-42.2	89.1
	3 rd	0	0.9994	80.84	76.02	-43.4	96.8
	4 th	0	0.9907	17.35	12.7	-45.9	42.35

TABLE 4: The thermodynamic parameters for the Cu(II) and Mn(II) complexes from DTA

Copmlex	Thermal nature process	t peak ^{0}C	ΔE KJ/mole	ΔH KJ/mole	ΔS J/mole	ΔG J/mole
A $[CuHL^1(H_2O)_3].2H_2OEtOH$	Exothermic	209-820	80	75	-88	583
C $[Cu_2 MnL^2(H_2O)_8].4H_2O$	Exothermic	241-566	73	58	-130	1290
D $[CuHL^3(H_2O)_3].\ H_2O$	Exothermic	184-534	199	196	-117	319
F $[Cu_2 MnL^4(H_2O)_8].8H_2O$	Exothermic	250-429	177	173	-46	159

cept and linear slope of such stage the A and E* values respectively were determined. The other thermo-kinetic parameters (ΔH*, ΔS*, ΔG*) were calculated using the relationshi ps:

$\Delta H^* = \Delta E^* - RT$

$\Delta S^* = R\,[In\,(Ah/KT)\,-1]$

$\Delta G^* = \Delta H^* - T\Delta S^*$

where K is the Boltzmann constant and h is the Planck's constant. The order and thermo-kinetic parameters of the chosen complexes are listed in (TABLE 3).

2. DTA analysis

The DTA curves for complex A showed three endothermic peaks at (59-99°C) (99-169°C very small) and (169-209°C) due to loss of lattic and coordirated ethanol and water molecules. The exothermic peak appearing at (209-820°C) is related the decomposition of the organic part of complex and formation to CuO. The DTA curves of complex C showed an obvious exthromic peak at (150-241°C) related to loss of coordinated water molecules and an other exothermic at (241-566°C) due to decomposition of the ligand in the complex leading to the formation of copper and manganese oxides.

The DTA Curves for complexes D and E showed two endothermic peaks at (55-92°C, 184°C and 72°C, 133°C for complexes D and E respectively, which are related to loss of lattic and corrdinated water molecules. Also exothermic peaks are observed within the (184-534°C) and (250-429°C) ranges due to decompostion of complexes and formation of the metal oxides.

The thermodynamic parameter for the complet decomposition steps are calculated according to the following equation:

$\ln K = \ln A - E\alpha/RT$

k is the reaction constant, A is the frequency factor, R = gas constant, and E_a the activation energy. Thus ln k will vary linearly with 1/T. This plot yields E_a from the slop.

Also[20,21]

$$In\left(\frac{K}{T}\right) = In\left(\frac{K_B}{h}\right) + \frac{\Delta S}{R} - \frac{\Delta H}{RT}$$

where K_B is Boltzmann's constant; h Planck's constant, R the gas constant. A plot of lnk/T vs.1000/T will be linear; the slope yields Δ H. The intercept, after allowance for in k_B/h=23.76, provided the volues of ΔS.The Gibbs free energy can be calculated from the equation.

$\Delta G = \Delta H - T\Delta S$

The data obtained are collected in (TABLE 4).

The data of thermal andysis showed that the addition of Mn(II) to the Cu(II) complexes decreased the stability of complexes. Also, the nature of ligand showed an effect on the thermal stability. The complex with COOH group ligand is more stable than complex of ligand with OH group. The thermal stability of the complexes can be arranged in the following order: A>C>D>E

3. IR-spectra

1. The spectrum of the complex A exhibited the band due to $\nu_{symN=N}$. at 1550cm^{-1} which was shifted to lower wave number than for the free ligand (1579cm^{-1}) indicating that this group contributed to chelation with Cu(II) .The position of $\nu_{symN=N}$ was shifted to 15470cm^{-1} for comples B and C.
2. The spectrum of ligand H_3L^1 and complex A showed a band at 1621cm^{-1} which due to C=O of the formyl group.This band disappeared from the spectra of complexes B and C with the appearance of a new band for $\nu_{C=N}$ at 1594cm^{-1} in case of complex B which was shifted to 1588 cm^{-1} for complex C, indicating that C=N graups contributed to complexation with Mn (II).
3. The band due to the phenalic-OH groups of the ligand H_3L^1 at 1328cm^{-1} was shifted to 1385cm^{-1} for complex A.The phenalic-OH groups for complexes A and B (1385cm^{-1}) disappeared from the spectra of complex C indicating that these groups contributed to chelation with the copper and manganese ions.
4. The spectrum of the complex D exhibited a band due to $\nu_{symN=N}$. at 1575cm^{-1} which was shifted to lower wave number than for the free ligand (1583cm^{-1}) indicating that this group contributed to chelation with Cu(II) .The position of N=N shifted to 1550cm^{-1} for complexes E and F
5. The spectrum of ligand H_3L^3 and complex D showed a band at 1610cm^{-1} which is due to C=O of the formyl group.This band disappeared from the spectra of complexes E and F with the appearance of a new band for νC=N at 1580cm^{-1} in case of complex E which was shifted to 1600cm^{-1} for complex F, indicating that C=N graups contributedin complexation with Mn (II).
6. The band due to the phenalic-OH groups of the ligand H_3L^3 at 1326cm^{-1}was shifted to 1300cm^{-1} for complex D.The band of phenalic-OH groups for complexes D and E(1300cm^{-1}) disappeared from the spectra of complex F indicating that thesegroups contributed to chelation with the copper and manganese ions.
7. The band for Mn-O appeared in the spectra of complexes C and F at 511cm^{-1} and 500cm^{-1} respectively. The spectra of all complexes display two new bands situated at 560-538cm^{-1} and 477-450cm^{-1} which are due to M-O and M-N stretching modls respectively[22].

Electronic absorption spectra

The electronic absorption spectra of the complexes under investigation were studied as nujol mulls and DMF solution; the λ_{max} values obtained are listed in(TABLE 5). The electronic absorptaion spectra of the compleses in DMF are comparable with those in nujol mull. However, it is found that the positions of the band due to d-d transition within the metal(II) ions exhibited some shifts to lower or higher frequency. The small shifts observed with some complexes are probably due to the changed medium refractive index and variation in its dielectric constant on going from the solid state nujol mulls to the solution in DMF.

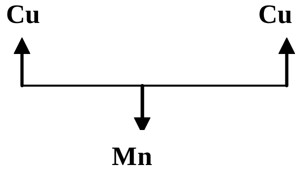
The spectrum of the mono and di Cu (II) complexes exhibited one absorption band at λ_{max} 440, 430, 360, 330nm for complexes A, B, D, E respectivitly due to $(^2B_{1g} \rightarrow {}^2T_{2g})$ transition indicating octahedral geometry around the Cu(II).

The d-d transition band for Mn(II) led to one absorption band at λ_{max} 340 nm and λ_{max} 310 nm for complexes C and F respectivity ,which can be assigned to the $(^6A_{1g} \rightarrow {}^4A_{1g})$ transition in octahedral geometry around the manganese ion.

Magnetic measurement

The magnetic susceptibilities of solid complexes were determined at room temp. using the Gouy method, from which the correspanding magnetic moments were calculated. Diamagnetic corrections were caried out using Paskal's constants; the data obtained are give in TABLE 1.

The observed magnetic moments for the Cu(II) complexes are much lower than the spin only values of one unpaired electron and indicated strong antiferromagnetic interaction between adjacent Cu (II) ions. For Cu and Mn in the same complex (C and F), the lower μ_{eff} values than the spin only value expected for the uncoupled Cu(II)-Mn(II)-Cu(II) unit, is due to bilateral exchange interaction in which perfect antiparallel alignment of spins on neighboring metal ions occurs[23]. The ground state may be schematized as:



The electron spin resonance spectra

The g_{eff} values calculated from X-band ESR spectra are recorded in TABLE 1.

The ESR spectra of the Cu(II) complexes A, B, D and E display a broad signal. The apparent broading of the ESR Signal can be dute to leteral antiferromagnetid interaction of Cu-Cu ions which is in agreement with the data of magnetic movement measurments.

The ESR spectra of Cu-Mn complexes (C and F) exhibit 2 sigenals at g_{ef} (1.82, 1.84) for Cu ion and (1.65 and 1.67) for manganese ion for complexes C and F respectively.

TABLE 5 : The data of electronic absorption spectra for complexe

Complex	d-d transition Nujo mull (λ_{max} nm)	d-d transition DMF (λ_{max} nm)	Assignment
A $[CuHL^1(H_2O)_3].2H_2OEtOH$	491	430	$^2B_{1g}\to{}^2T_{2g}$
B $[Cu_2HL^2(H_2O)_6].3H_2O$	479	430	$^2B_{1g}\to{}^2T_{2g}$
C $[Cu_2MnL^2(H_2O)_8].4H_2O$	479 415	430 410	$^2B_{1g}\to{}^2T_{2g}$ $^6A_{1g}\to{}^4A_{1g}$ (Mn II)
D $[CuHL^3(H_2O)_3].H_2O$	479	360	$^2B_{1g}\to{}^2T_{2g}$
E $[Cu_2HL^4(H_2O)_6].5H_2O$	-	330	$^2B_{1g}\to{}^2T_{2g}$
F $[Cu_2MnL^4(H_2O)_8].8H_2O$	461 415	340 310	$^2B_{1g}\to{}^2T_{2g}$ $^6A_{1g}\to{}^4A_{1g}$ (Mn II)

The shape of ESR signals reflects the lateral exchange interaction this indicated spin structure where all the S_{Mn} = 5/2 local spins are aliganed along the same direction for the S_{Cu}=1/2 local spins are aligned along the opposite direction. Also the negative contribtion to the g_{eff} values related to that of the free electron (2.0023) reflects a decrease of the covalent character of the bonding between the metal ion and the ligand molecule[24].

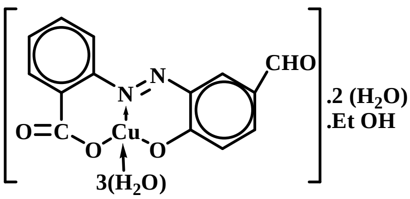
The shape of the signal and g_{eff} values indicate octahedral geometry of the ligand around cupper and manganese ions. Also, it reflects that the metal ion is present in a nonhomogenous field.

Based on the results of elemental and thermal analysis, the IR, UV/Vis and ESR spectra, magnetic mmomment determination, the complexes involved in the present study can be formulated as follows:

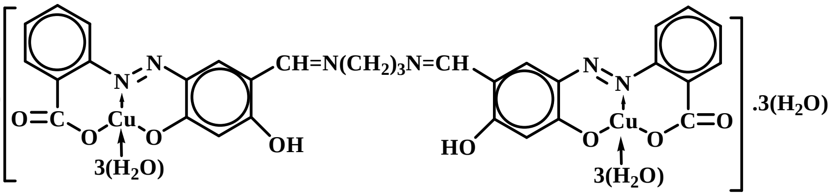
TABLE 6 : Antimicrobial effects of ligands and their complexes

Compound conc. 0.1 %	Microorganisms (inhibition zone/cm) *Bacillus subtilis*	*E.coli*	*Staphylloccus aerues*	*Pesudomonas auregonosa*	*Salmonella SPP.*
HL^1	-	-	-	-	-
HL^3	-	-	-	-	-
A $[CuHL^1(H_2O)_3].2H_2OEtOH$	0.3	3.1	2.8	3.6	2.5
B $[Cu_2HL^2(H_2O)_6].3H_2O$	1.9	2.0	2.1	1.7	1.3
C $[Cu_2MnL^2(H_2O)_8].4H_2O$	1.5	1.8	2.2	2.2	2.1
D $[CuHL^3(H_2O)_3].H_2O$	2.7	1.9	2.2	2.6	1.3
E $[Cu_2HL^4(H_2O)_6].5H_2O$	2.8	2.5	2.1	2.9	2.3
F $[Cu_2MnL^4(H_2O)_8].8H_2O$	2.9	2.8	3.0	2.6	2.7

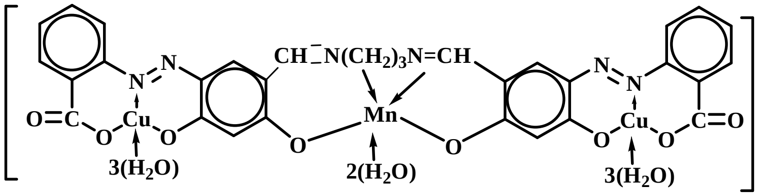
(-) reveal that the compounds have not any activity against to the microorgnisms



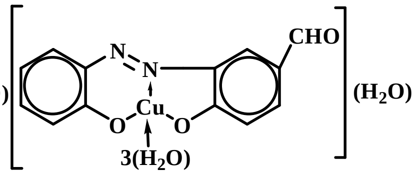
Complex A



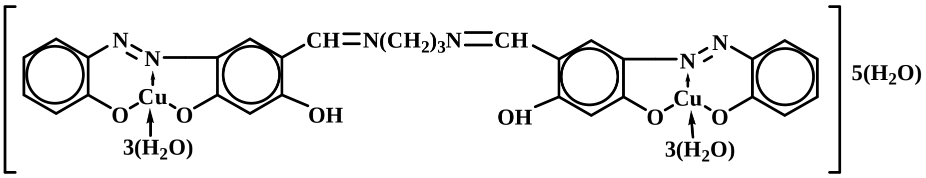
Complex B



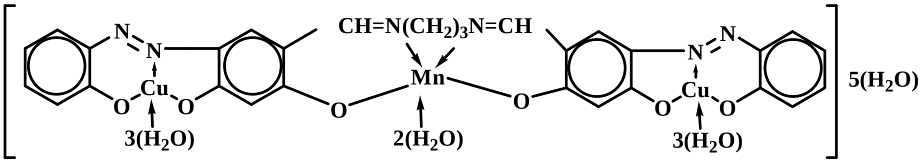
Complex C



Complex D



Complex E



Complex F

Biological activity

Bactericidal activities of the ligands and their metal complexes against pathogenic bacterial strains (clinical isolate) are recorded in TABLE 6 It has been suggested that the ligands with N and O donar system might inhibited enzymes activity, since enzymes which require free hydroxy group for their activity appear to the especially susceptible to deactivation by the ions of the complexes. The complexes facilitate their diffusion through the lipid layer of membranes to the sites of action ultimataly killing them by combining with -OH groups of certain enzymes.

The variation in the effectiveness of different biocidal agent against different organisms[25] depends on the impermeability of the cell. The hydrocarbon acts as a lipophilic group[26] to drive the compound through the cell. Chelation reduces the polarity of the central ion mainly because of the partial sharing of its positive charge with the donor groups and possible π-electron delocalization within the whole chelate ring.This chelation increases the lipophilic nature of the central atom which favours its permeation through the lipid layer of the membrane.

In this study, the results show that the all compounds exhibited inhibitions towards all tested organism excepte ligands under the test conditions TABLE 6.This reflect that the possible use of the metal complexes as a good antibacterial agents for the strains envolved in the present study.

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