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SPECTRAL, THERMAL, X-RAY DIFFRACTION AND ANTIMICROBIAL STUDIES OF SOME BIVALENT METAL CHELATES OF JUGLONE

A. B. PAWAR, S. R. BAMANE[a], K. D. JADHAV and R. G. SARAWADEKAR[*]

Department of Chemistry, Bharati Vidyapeeth Deemed University, Yashwantrao Mohite College, PUNE – 411038 (M.S.) INDIA
[a]Metal Oxide Research Laboratory, Dr. Patangrao Kadam Mahavidyalaya, SANGLI – 416146 (M.S.) INDIA

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ABSTRACT

Five metal chelates of the type M $[Jg]_2$ where M = Zn, Cd, Hg, Sn and Pb and Jg = 5-hydroxy-1, 4-naphthoquinone (Juglone) have been synthesized. All chelates have been characterized by modern methods such as elemental analysis, FTIR, X- ray diffraction, Electronic spectra, Thermogravimetry, Differential thermal analysis and electron microscopy with EDAX analysis. All chelates are found to be colored. These chelates are thermally stable up to 500^{o}C and all are crystallite in nature and the crystallographic parameters are calculated, which shows that these crystals belong to triclinic. The particle size of Jg, Zn-Jg and Pb-Jg is 104, 52 and 39 nm, respectively. The ligand and the metal chelates have been screened for antimicrobial activity on Gram positive, Gram negative and antifungal activity and results are compared with standard cisplatin compound. The results are encouraging.

Key words: Juglone, Metal chelates, IR, NMR, SEM, Antimicrobial activity, Electronic spectra.

INTRODUCTION

The spectral and thermal properties of juglone chelates with divalent Cu, Zn, Co, Ni and Pb were reported by Bottei and Mc Eachern[1] The dissociation constants of Ni and Zn juglonates have been reported by Kopterivo et al.,[2] IR and EPR studies on Ni juglone salts have been reported by Aizenberg et al.,[3] Juglone gave sharp and sensitive colour changes at the end point when used as indicator for the determination of Cu (II), Co (II), Ni (II), and Mg by complexometric titration with EDTA[4]. Kinetic parameters were determined for decomposition of unhydrous Cd, Y (III), and Sm (III) juglone complexes. Hydrated complexes lose H_2O at 50-125^{o}C and air oxidation occurs at 50- 900^{o}C for unhydrous complexes[5]. Stability constants and thermodynamic parameters were reported for Cd (II) juglone salt at 20 and 40^{o}C and it was reported that all the system have positive entropies[6]. Fe (II) complex of juglone was synthesized and IR spectra were discussed along with electronic spectra, which resulted that it formed 6 member chelates rings[7]. Proton NMR and ^{13}C NMR of zinc juglonate, along with TG/DTA studies of Zn Cu and Ni juglonate were reported by Lalia Kantouri and Ballola-Christianopoulou[8]. Mn (II) juglone complex was prepared and tested *in vitro* against Bacillus species and *Staphylococcus aureus* and reported that the metal complex displayed more antibacterial activity than the ligand. Also the chelates had a strong selective activity towards B. Stereo thermophilus[9]. Kinetic parameters such as energy of activation (E) and

Available online at www.sadgurupublications.com
[*]Author for correspondence; E-mail: rgsarawadekar@yahoo.co.in; Ph.: 020-25433383; Fax: 020-25440201

prier of reaction (n) of Cd $(C_{10}H_5O_3)_2$ have been reported but the temperature zone 150-900°C[5]. Juglone undergoes sensitive colour reaction with Ni (II) and Co (II) and fairly sensitive reaction with Zn (II) and Mn (II) at pH 6.8. All complexes had a 1 : 2 ratio for metal to ligand (II)[10]. Formation constants of the Cu (II), Ni (II), Co (II), Zn (II), Cd (II) and Mn (II) complexes of juglone have been reported by Kido et al.,[11] Co (II) complexes of juglone was prepared and found that it contain 1.5 H_2O water of crystallization with intense dark violet colour precipitates[12]. Though some was is reported on metal complexes of juglone, still there is a scope to work on these complexes. In this paper, we report metal complexes of zinc, cadmium, mercury, tin and lead of juglone. The characterization includes XRD, MID IR, FAR IR, DTA/TG analysis, electronic spectra, SEM & EDAX data and antimicrobial activity against microorganisms such bacterial and fungal.

EXPERIMENTAL

Materials and methods

The ligand juglone (5-Hydroxy-1, 4-napthaquinone) was synthesized in laboratory by using the method describe earlier in our paper[13]. Silver studied of Zn (II), Cd (II), Hg (II), Sn (II) and Pb (II) chelates were prepared by using AR grade chemicals. The standard compound Cisplatin is obtained from Naprod Life Sciences Pvt. Ltd. Boisar, Thane (India).

Preparation of metal chelates

The metal chelates were prepared as follow: In 100 cm^3 of warm water moles of metal salts were dissolved. To this solution 1.00 g (5.8 mmoles) of 5-Hydroxy-1, 4-napthaquinone (Juglone) dissolved in 100 cm^3 of hot methane were added slowly under continuous magnetic stirring. The pH was adjusted to 5.0-6.00 with 25% NH_3 ammonia.

The mixture was left stirring at 20°C temperature for 3 hr and then was kept in refrigerator overnight. The precipitate were washed several times with water ethanol and finally with diethyl ether and was added.

Instrument analysis

The MID IR spectra recorded in KBr matrix an JASCO FTIR and FAR IR spectra were recorded on Nicolet 6700 diamond ATR model in the range of 700-100 cm^{-1} using powder sample. Elemental analysis was carried out with a Perkin Elmer 2400 series for C, H, O and N. Electronic spectra were recorded on JASCO 530 model in KBr matrix and methanol solution. DTA/TGA curves were taken on Shimatzu 60 H model using 10 electron/mm heating rate 800°C maximum temperature in air.

Scanning electron microscopy was used to get images of these chelates on Vega II SB model and EDAX analysis of element was carried out on OXFORD INCA PENTA with TESCAN VEGAII SB.

X-ray diffraction patterns were obtained on Bruker D advanced diffracto meter at room temperature.

Antimicrobial activity testing

Test organism the antimicrobial activity of lignad metal salts and synthesized metal chelates of transition metals Zn (II), Cd (II), Hg (II), Sn (II) and Pb (II) against bacteria *Escherichia Coli* (NCIM 2065). *Bacillus subtities* (NCIM 2063) *Sstaphylococcus aureus* (NCIM 2079) *Candida albicans* (NCIM 3471) and *Klebsiella pneumoniae* strains collected from National Chemical Laboratory, Pune.

Maintenance of culture

The cultures of bacteria fungi were maintained on Nutrient agent muller Tinton Agar and Subcultured accordingly.

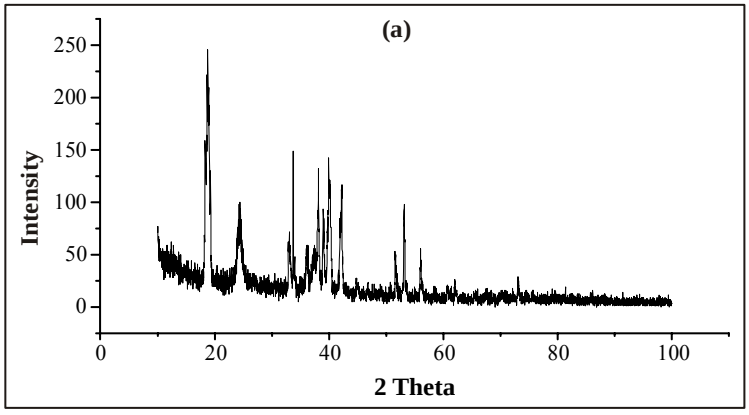
Plate diffusion method

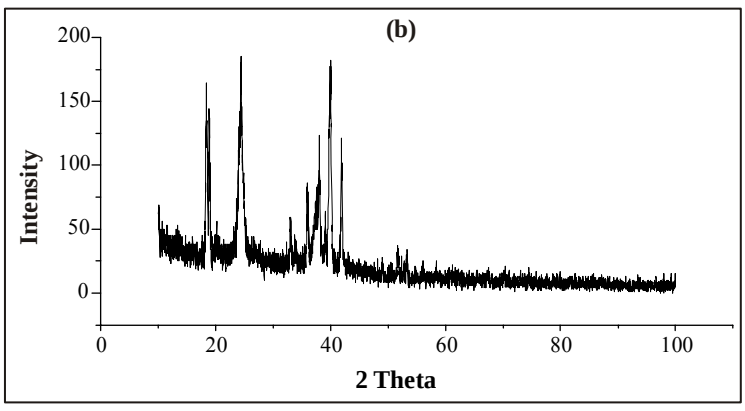
The 100/µL cell suspension (10^8 cell/ mL) of bacteria and 100/µL of spore suspension of mold were spread on the n Agar for bacteria and mueller Hinton agar for fungi. Then wells were bored in the media in which DMSO (solvent) ligand metal salts and metal chelates solutions were poured for each organisms. The petri plates were then incubated at 370°C for 48 hrs for bacteria and 30°C for 5 days for fungi. All the aseptic operations were carried out under the laminar air flow unit. The genes of inhibition were measured in mm.

RESULTS AND DISCUSSION

X-ray diffraction

The crystal structure of Juglone was presented by Chadwick and Hall[14] and reported that Juglone crystals were monoclinic a = 7.34, b = 7.69, c = 13.91, β = 99.20. Later Welton and McCarthy[15] reported that the crystals structure of Juglone exhibits as monoclinic with space group $P_{2\ 1/n}$ (14) with a = 13.912, b = 7.7112, c = 7.3201, β = 98.905^0. We have synthesized Juglone[13] and x-ray diffraction was carried out using Cr Kr 2.28970 A° in the range of 10 to 80°. X-ray diffraction patterns of juglone, zinc juglonate and lead juglonate have been shown in following :





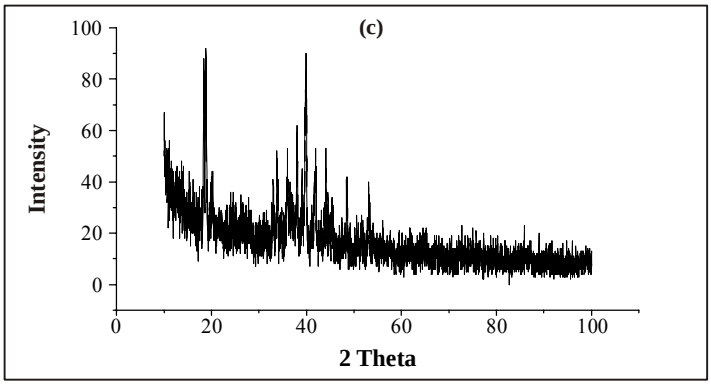


Fig. 1: X-ray diffraction patterns of (a) Juglone, (b) Zinc juglonate and (c) Lead juglonate

The powder diffraction data was computed using McMaille V 4.00 (16) and found that Juglone belongs to monoclinic group and h k l data in given in Table 1.

Table 1: Miller indices and 2 theta values of Jg

S. No.	h	k	l	TH (OBS)	TH-ZERO	TH (CLAC)	DIFF
1	1	0	-1	18.805	18.785	18.794	-0.009
2	0	0	1	18.990	18.970	18.958	0.012
3	0	2	0	33.655	33.635	33.639	-0.005
4	1	2	0	36.045	36.025	36.025	0.000
5	3	0	-1	36.335	36.315	36.305	0.010
6	2	0	-2	38.125	38.105	38.118	-0.013
7	3	0	0	38.190	38.170	38.186	-0.016
8	1	1	-2	39.955	39.935	39.933	0.002
9	3	1	-1	40.215	40.195	40.180	0.014
10	2	1	-2	41.870	41.850	41.852	-0.002
11	0	1	2	42.196	42.176	42.170	0.006

From this data it is observed that the cell constants are a = 11.1757, b = 7.9095, c = 7.3994 A^o, $\beta = 110.12^0$, which is all together new. It is noticed that the formation of juglone sturucture is varying in nature, as layer by layer growth of crystals is varying.

In the case of zinc juglonate (Zn-Jg) the h k l data is shown in Table 2.

Table 2: Miller indices and 2 theta values of Zn-Jg

S. No.	h	k	l	TH(OBS)	TH-zero	TH(CLAC)	DIFF
1	0	0	1	24.440	24.405	24.410	-0.005
2	1	1	-1	32.970	32.935	32.944	-0.009
3	2	2	0	35.925	35.890	35.871	0.019

Cont…

S. No.	h	k	l	TH (OBS)	TH-zero	TH (CLAC)	DIFF
4	0	2	-2	38.165	38.130	38.120	0.010
5	1	-2	1	39.095	39.060	39.059	0.001
6	2	2	1	39.770	39.735	39.741	-0.006
7	2	3	0	40.030	39.995	40.005	-0.010
8	0	3	-2	41.900	41.865	41.868	-0.003
9	0	3	0	48.950	48.915	48.892	0.023
10	1	-2	0	51.745	51.710	51.720	-0.010
11	0	4	-1	53.285	53.250	53.259	-0.009

The calculated data suggested that these crystals belong to triclinic group. The cell constants are a = 13.8414, b = 6.7297, c = 7.0709 A^o, $\alpha = 82.098^0$, $\beta = 117.013^0$, $\gamma = 71.570^0$ and volume 525.006 (A^o) 3. The calculated density is 1.660 g/cm^3 and the group is space group H-M symbol P1. The h k l data of lead juglonate (Pb-Lw) is presented in Table 3.

Table 3: Miller indices and 2 theta values of Pb-Jg

S. No.	h	k	l	TH(OBS)	TH-zero	H(CLAC)	DIFF
1	0	1	0	18.365	18.349	18.369	-0.020
2	1	1	0	18.785	18.769	18.743	0.026
3	0	2	-1	32.970	32.954	32.962	-0.008
4	1	0	3	33.850	33.834	33.828	0.006
5	1	2	-2	35.985	35.969	35.989	-0.020
6	2	2	0	38.015	37.999	38.012	-0.013
7	0	1	2	39.010	38.994	38.990	0.004
8	2	0	1	39.895	39.879	39.866	0.013
9	2	0	3	41.910	41.894	41.893	0.001
10	1	2	2	44.145	44.129	44.131	-0.002
11	1	-2	2	45.410	45.394	45.385	0.009
12	2	3	-1	48.455	48.439	48.419	0.020
13	3	1	2	53.245	53.229	53.245	-0.016

The crystals of lead juglonate belongs to triclinic group and suggested cell could be a = 7.7985, b = 9.1353, c = 12.1794A $\alpha = 103.958^0$, $\beta = 70.008^0$, $\gamma = 62.99^0$. The volume of crystal would be 639.839 $(A^o)^3$ and the calculated density is as 1.958638 g/mL. Each crystal is made up of 2 units i.e. = 2. Its space group is H-M symbol P-1 and empirical formula is $C_{40}H_{2O}O_{12}Pb_2$.

Thermal analysis (TG/DTA)

Thermogravimetry (TG)

Juglone: TG/DTA of Juglone was carried out in air and heated up to 800^{o}C with heating rate as 10^{o}C/min. TG curve shows one step starts at 112.79^{o}C to 185.74^{o}C and the weight loss up to 20.25% was recorded. TG curve is shown in Fig. 2. The second step is slow but continuous weight loss is

observed in the range of 287°C to 387.17°C and weight loss is found to be 22.2%. The sample is heated up to 800°C, which shows continuous weight loss and finally it gives 44.80% residue.

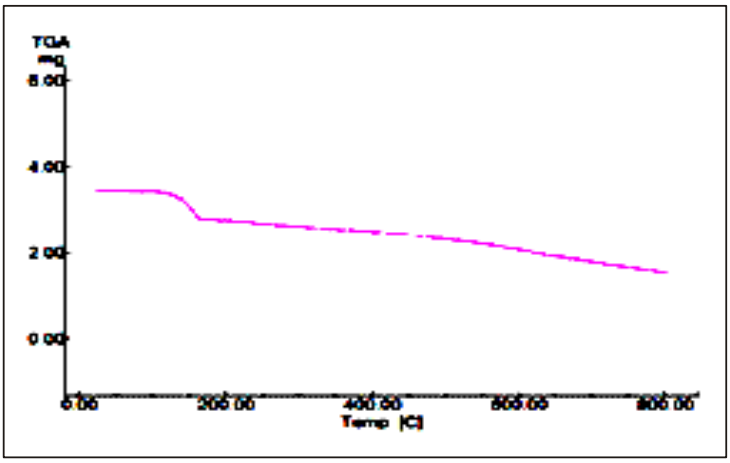


Fig. 2: TG curve of juglone

Zn-Jg (Zinc juglonate) Zn $(C_{10}H_5O_3)_2$

Fig. 3 shows TG curve of Zn-Jg. TG curve shows one sharp weight loss in the temperature range 110.89 to 200.66°C and it gives weight loss equal to 17.79%. It is similar to Juglone decomposition. Then the weight loss is continued up to 800°C and finally it shows 46.35% weight loss. The mechanism can be explained as -

$$Zn\ (C_{10}H_5O_3)_2 \longrightarrow Zn\ (C_{10}H_2O_2)$$

$$Zn\ (C_{10}H_2O_2) \longrightarrow ZnO + 9C$$

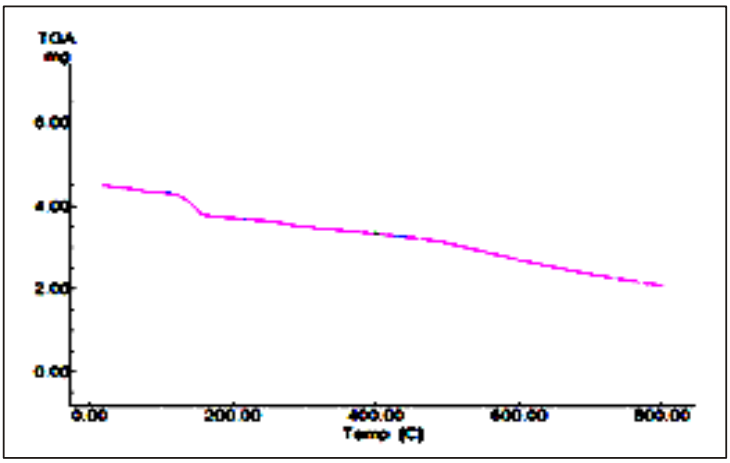


Fig. 3: TG curve of Zn (II) juglonate

Pb-Jg (Lead Juglonate) Pb $(C_{10}H_5O_3)_2$

TG curve of lead juglonate shows weight loss in two steps as shown in Fig. 4. First step is in the temperature range 110.90 to 150.57°C and the second between 540.85 to 650°C is found. The first step, weight loss is about 9.8% and the second one is 80%. The end product is lead metal or metal oxide as residue.

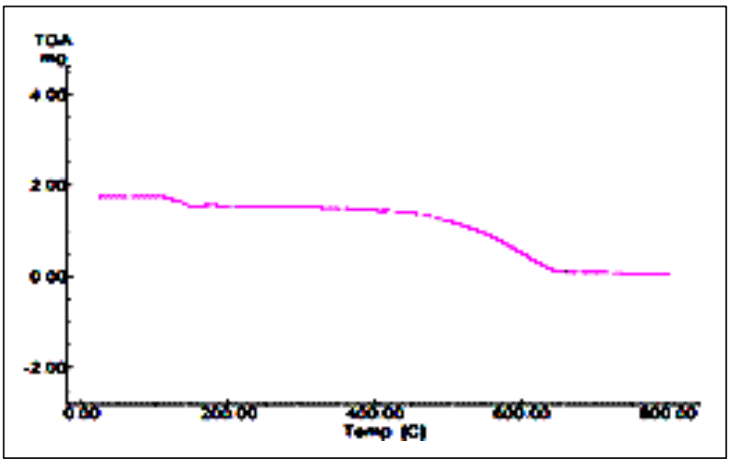


Fig. 4: TGA Curve of Pb (II) Juglonate

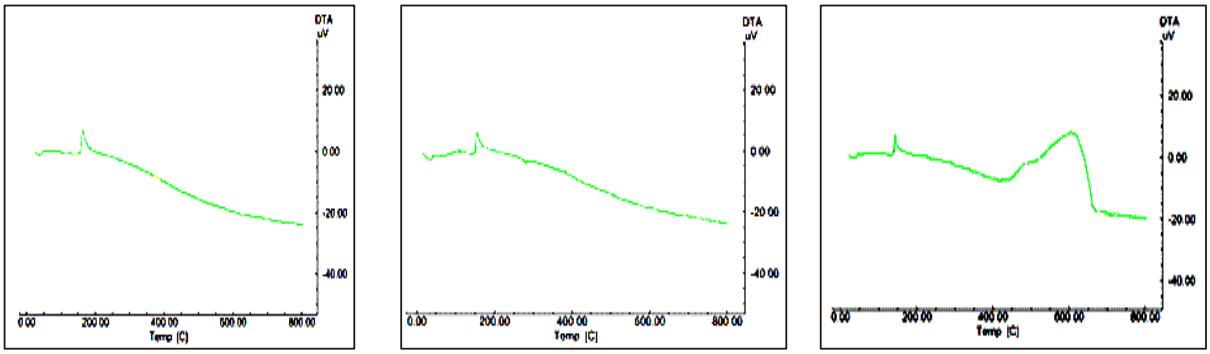
Differential thermal analysis (DTA)

Fig. 5 shows DTA curves of Jg, Zn-Jg and Pb-Jg.

1. Jg (Juglone): From the DTA curve, it is noticed that only one exothermic peak is observed. The peak temperature is at 163.72°C and in the range of 157.84 to 195.81°C. It is attributed to the decomposition of Juglone.

2. Zinc Juglonate: This chelate shows only one exothermic peak with peak temperature range at 149.47 to 201.84°C. This is attributed to the decomposition of Juglone which can be compared with decomposition of Juglone alone

3. Lead Juglonate: This chelate shows two exotherms, first is due to decomposition of Juglone which is similar to juglone. The second exotherm is due to further decomposition of the remaining moiety. The first decomposition peak temperature is at 143°C in the range of 136.82 to 229.67°C and the second exothermic peak is observed at 605.50°C in the temperature range 527.17 to 638.73°C.



Jg Juglone **Zn-Jg** **Pb-Jg**

Fig. 5: DTA curves

Kinetics from TG data

Kinetic parameters were calculated by using computer code developed by Sarawadekar et al.[17] The decomposition of Juglone, in the temperature range 112.9 to 185.74°C was carried out and also for second stage calculations were carried out. The data is presented in Table 4.

Table 4: Kinetic parameter for decomposition of Jg (Juglone)

S. No.	Parameters	Step I	Step II
1	Temp range	112.79 -185.74^0	287.54 -387.17 ^{0}
2	n order of reaction	1	1
3	Ea Energy of activation	15.15 kJ/mol	50.14 kJ/mol
4	R Regression coefficient	0.9587	0.993
5	Log (A) Frequency factor	3.4381	2.027
6	S Entropy of activation	-194.8 J/mol/ K	-232.12 J/mol/ K
7	G Free energy of activation	206.34 KJ/mol	196.46 KJ/mol
8	H Enthalpy of activation	119.73 KJ/mol	49.71 KJ/mol

The thermal decomposition kinetics from TG data was computed and the data is given in Table 5 for Zn-Jg (zinc juglonate) and Pb-Jg (lead juglonate).

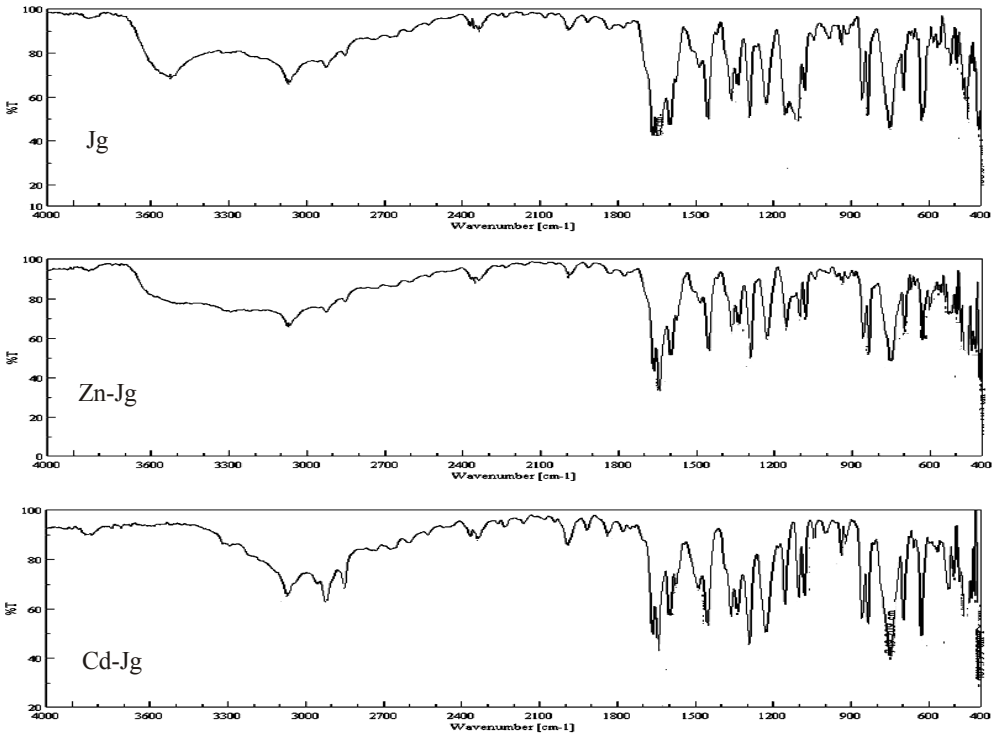
Table 5: Kinetic parameter for decomposition of Zn-Jg (zinc juglonate) and Pb-Jg (lead juglonate)

S. No.	Chelate	Ea KJ/mol	n	Log (A)	TG step	R
1	Zn-Jg	28.68	1	1.6184	I	0.9939
2	Pb-Jg	150.23	1	8.9138	I	0.9647
3	Pb-Jg	70.00	1	2.3983	II	0.9939

Where Ea = Energy of activation; n = order of reaction;
Log (A) = Frequency factor; R = Regression coefficient

I.R Spectroscopy

The I.R spectra of Jg (Juglone) and its chelates are given in Fig. 6.



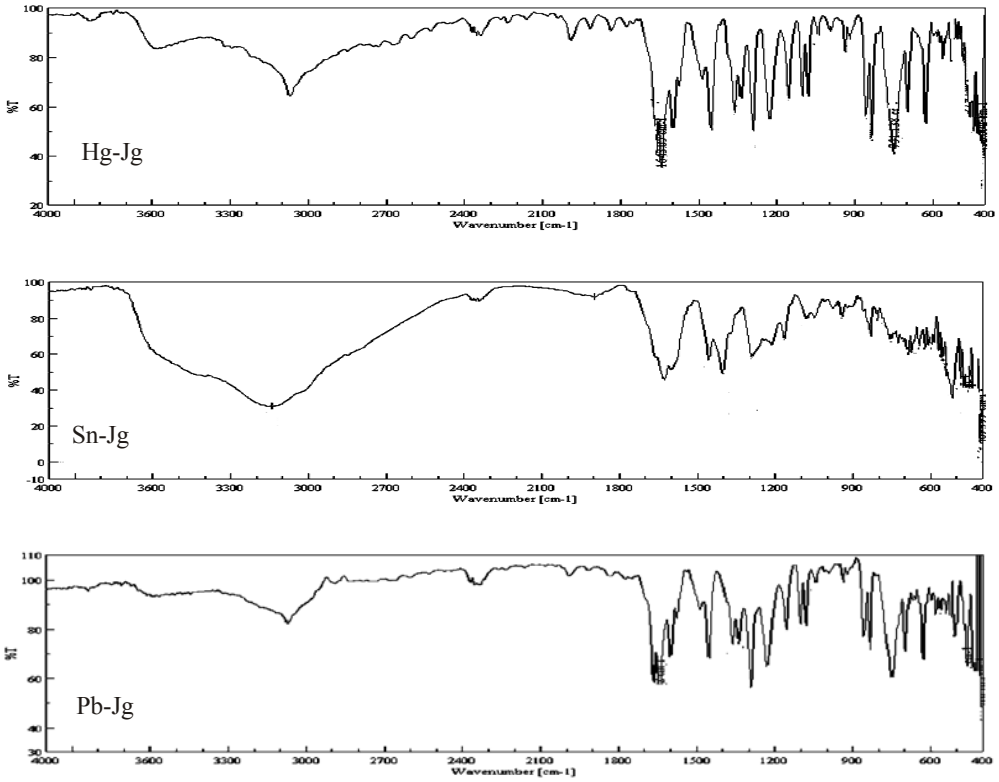


Fig. 6: MID IR spectra of Jg and its chelates

The zinc, cadmium, mercury, tin and lead chelates show absorption in the range of 3073-313 cm^{-1} indicative of C-H Stretching mode. Only Cd-Jg shows peak at 3525 cm^{-1} for OH stretching mode. The zinc, mercury, tin and lead chelates do not show strong absorption in thin region and anhydrous as seen Table 6. The free carbonyl and hydrogen bonded carbonyl are assigned to 1665 and 1643 cm^{-1}, respectively. Kido et al.[11] assigned these absorptions at 1670, 1647 cm^{-1}, respectively. The metal chelates show many strong bands in 1700 to 1000 cm^{-1}, with their spectra similar to that of Jg (Juglone) itself. These would be due to chelated carbonyl stretching mode, relative shifts due to chelation may be determined for chelates. The amount of the chelated carbonyl group of the Jg (juglone) chelate can be compared. The shift of these are varying in the rang 130-142 cm^{-1}. The maximum shifting is shown by Zn-Jg chelate. The other important peak is C-O stretching which is observed at 1228 ± 1 cm^{-1}. It is observed that these peaks are well resolved.

Table 6: Characteristics IR (cm^{-1}) bands of Jg and its metal chelates

S. No.	Compound	OH	C=O	C=O	C-O
1	Jg	--		1665	1228
2	ZnJg	--	1599	1665	1229
3	CdJg	3525	1600	1665	1227
4	HgJg	--	1600	1665	1227
5	PbJg	--	1629	--	1227
6	SnJg	--	1600	1665	1227

UV-VIS Spectroscopy: (UV-VIS)

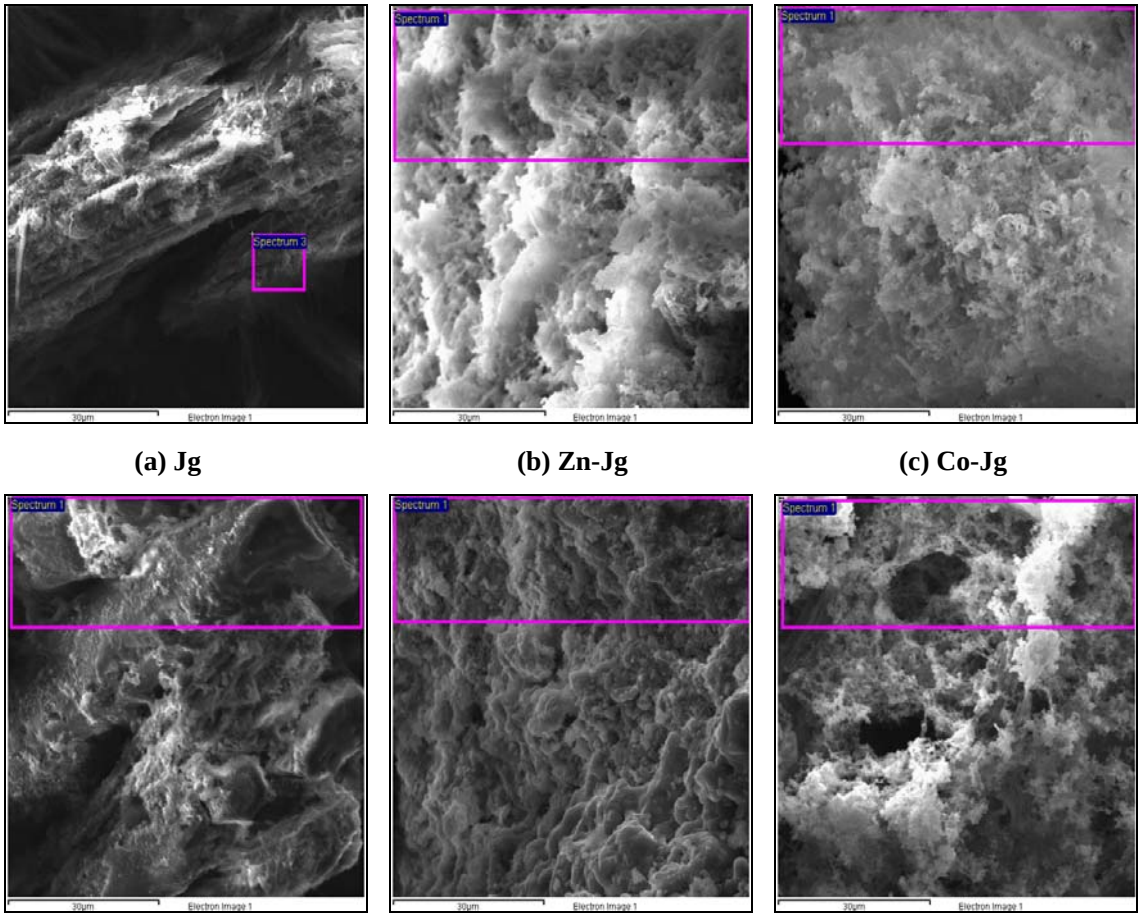
The electronic spectra (UV-VIS) spectra of Jg (Juglone) and its metal chelates were recorded in solid state and in solutions. The data in presented in Table 7. In solid state it shows all three transitions i.e. benzenoid, quinonoid and n to π*. In solid state the first transition shows variations in absorption band with some shifts. Generally in solutions, quinonoid transition absorption is not resolved. Only Sn-Jg chelate shows peak at 342 nm in methanol. The absorption band at 420 ± 8 nm shows good relation and it is attributed to absorption to n → π* transition. In the solid state the absorption is observed at higher wave lengths.

Table 7: Electronic spectra (UV-VIS) of juglone and its metal chelates

S. No.	Compound	Principle Band Wavelength					
		BET		QET		$n \rightarrow \pi^*$	
		Solid	Solution	Solid	Solution	Solid	Solution
1	Jg	288	248	344	-	414	406
2	Zn- Jg	247	248	307	-	420	406
3	Cd- Jg	289	248	351	-	496	406
4	Hg- Jg	246	248	348	-	423	404
5	Sn- Jg	248	234	337	342	448	-
6	Pb- Jg	289	250	325	-	423	406

Metal organic framework

The metal organic frame work (MOFS) is fascinating class of solid state inorganic-organic hybrid materials. Research in to this compound is expanding very rapidly owing to their exciting combination properties for advanced functional materials in gas separation & storage, chemical sensing, catalysis as well as medical applications. These compounds widely used chemotherapy agents. All synthesized metal organic frame works have shown more inhibitory activity against bacteria, yeast and fungi as compared to parental ligands. The antimicrobial activity explores on the basis of overtone concept of cell permeability. Fig. 7 shows SEM pictures of Jg (Juglone) and its metal organic frameworks. The growth orientation on layer by layer growth of some MOF required the coordination through ligand-metal ion which forms the two dimenational layer of the MOF. It was impeded by presence of oxime and hydroxyl groups. The material growth under co-ordination modulation was significantly enhanced compared that of the usually obtained microcrystalline powder of metal chelate. The growth of metal chelate of Jg (Juglone) is observed as bulk crystals. The crystalline size of the chelates is found to be nano range described earlier. This difference was attributed to the higher structural defect of usual materials as compared with co-ordination modulated nano–MOF. The characterization of well defined, stable self assembled mono layers (SEMS) of organic ligands at crystal face of MOF has not been documented to date. SEM pictures of Jg (Juglone) and its metal organic framework shows different structures. Form these SEM scanning, EDAX Analysis was carried out for Jg (Juglone) and MOFS. The data for % O, % C and % metal is shown in Table 8. It can be seen that the observed data and calculated data compared well except for metal contents which cannot be explained.



(a) Jg **(b) Zn-Jg** **(c) Co-Jg**

(d) Hg-Jg **(e) Sn-Jg** **(f) Pb-Jg**

Fig. 7: SEM photographs of (a) Jg, (b) Zn-Jg, (c) Cd-Jg, (d) Hg-Jg, (e) Sn-Jg and (f) Pb-Jg

Table 8: Elemental C, H, N and O analysis by EDAX of Jg and chelates

S. No.	Compound	Method	%C	%O	%H	%Metal
1	Jg	Obsd.	70.39	29.61	--	--
		Cald.	68.98	27.44	3.44	--
2	Zn-Jg	Obsd.	67.39	26.60	---	6.01
		Cald.	58.36	23.32	2.44	15.18
3	Cd-Jg	Obsd.	70.39	28.07	--	1.54
		Cald.	52.37	20.93	2.18	25.40
4	Hg-Jg	Obsd.	62.42	32.01	--	5.51
		Cald.	43.93	17.55	1.88	36.88
5	Sn-Jg	Obsd.	45.78	42.64	--	11.53
		Cald.	51.67	20.64	2.15	25.23
6	Pb-Jg	Obsd.	54.36	17.88	--	27.76
		Cald.	43.40	17.34	1.80	37.43

Antimicrobial activity studies

Antimicrobial scanning results

The Jg ligand and its metal chelates are screened for their antimicrobial activities against *Escherichia coli, Bacillus subtilis, Candida albicans, Staphylococcus aureus* and *Klebsiella pneumonia*. The testing against growth of micro-organisms was carried out by using well diffusion method employing Mueller Hinton Agar (MAH) and culture in nutrient broth in each case of micro-organisms. The concentration of ligand and its metal chelates were chosen as 10^{-4} M. The plates were incubated at 35°C for 24 hours in incubator. The clear zone of inhibition of growth for the organism was measured in mm and the data is given in Table 7. Dimethyl sulphoxide i.e. solvent used shows no inhibition for all organisms under studies.

Table 9: Antimicrobial activities of 5-hydroxy-1, 4-naphthoquinone (Jg) and its metal chelates (inhibition zone diameter in mm)

S. No.	Compound	*Escherichia coli*	*Bacillus subtilis*	*Staphylococcus aureus*	*Klebsiella pneumoniae*	*Candida albicans*
1	DMSO					
2	Jg	17	28	Nil	Nil	Nil
3	Zn-Jg	Nil	19	13	17	Nil
4	Cd-Jg	17	14	14	Nil	13
5	Hg-Jg	20	32	Nil	14	35
6	Sn-Jg	Nil	13	13	14	14.5
7	Pb-Jg	16	19	Nil	16	12
8	Cisplatin	18	13	20	No test	Nil

All the metal chelates showed microbial activity against all organisms studied in this work.

The metal chelates show good activity against most of them organisms. The inhibition of the micro-organisms growth for metal chelate was found to be in the following order for *Bacillus subtilis*.

Hg-Jg > Zn-Jg & Pb-Jg > Cd-Jg > Sn-Jg & Cisplatin

The studies demonstrate that metal chelation can increase the antimicrobial activity than metal free ligand. It is responding that metal chelation reduce the polarity of the metal ion mainly due to partial sharing of its positive change with the donor group and possibly the δ electron delocalization occurring within the whole chelate ring system formed during co-ordination and results in increase of the lipophilic nature of the central metal atom[18]. It favors for its penetration through the lipoid layer of the membrane.

The transition metal chelates possess high degree of inhibition which can be due to the greater number of δ electrons which increase the electrostatic field around the metal ion. The results are better than standard cisplatin compound.

CONCLUSION

The metal chelates are thermally stable up to 500°C, which is a unique characteristic property. All these metal chelates are crystalline in nature and generally belongs to triclinic while juglone is monoclinic.

The coordination ability of ligand Jg towards M (II) chelates were examined by different spectroscopic methods that unequivocally determine the coordination sites of ligands Jg. It is observed that the ratio of the metal chelates is 1 : 2 for chelates of Zn, Cd, Hg, Sn and Pb. This data is supported by analytical work. Biological activity screening proved the good antimicrobial activity of ligand Jg (Juglone) and its metal chelates. The antimicrobial activity explored on the basis of overtone concept of cell permeability.

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