IOSR Journal of Pharmacy
e-ISSN: 2250-3013, **p**-ISSN: 2319-4219, www.iosrphr.org
Volume 2 Issue 6 ‖ Nov-Dec. 2012 ‖ PP.25-33

Synthesis, characterisation and antimicrobial activity of bivalent metal (Zn, Cd, Hg, Pb and Ag) chelates of 1, 2- naphthoquinone dioxime

[1]N. R. Gonewar, [2]V. B. Jadhav, [3]K. D. Jadhav, [4]S.S. Sakure,
[5]A. A. KILLEDAR and [6]R. G. Sarawadekar

[1,2,3,6]Bharati Vidyapeeth Deemed University, Pune (India)
Yashwantrao Mohite College, Pune 411 038
[4]S.B.B. alias Appasaheb Jedhe College, Shukrawar Peth, Pune 411002.
[5]Department of Chemistry, A Vishay Components India Pvt. Ltd., Loni Kalbhor
PUNE – 412201 (M.S.) INDIA

Abstract**—Transition metal chelates of the type M [NQO]$_2$ where M = Hg, Cd, Zn, Pb: NQO = 1, 2 - napthoquinone dioxime and Ag (NQO) have been synthesized. All chelates have been characterized by modern methods such as elemental analysis, FTIR, Electronic spectra. ^{1}H & ^{13}C NMR, Thermogravimetry, Differential scanning calorimetry and electron microscopy with EDAX analysis of chelates were carried out. Metal chelates of mercury, lead, zinc and cadmium are octahedral while silver chelate has been assigned square planer. These chelates are thermally stable up to 350^{0}C and all are crystalline in nature. Their particle sizes are in the range of 15-42 nm. The ligand and the metal chelates have been screened for antimicrobial activity on gram positive and gram negative bacteria and fungi and the results are compared with cisplatin as standard chemotherapy agent.***

Keywords**—1,2-naphthoquinone -2, oxime, X-ray diffraction, IR, NMR, SEM, Antimicrobial activity, Electronic spectra.***

I. INTRODUCTION

Our studies on 1-2-naphthoquinone-2, monoximes (1,2) and the metal chelates formed by them (3) have led us to investigate the vicinal dioxime compounds derived from aromatic nuclei. It is well known that the complex formation ability of the -C (C = NOH) – C (C = NOH) – is greatly influenced by the special arrangement of the oxime groups. The specific action of the grouping towards nickel ions has been attributed to the anti isomers of the compounds. A major aim to study such compounds originates from the fact that these types of ligands shows biological importance [4,10]. Oximes and dioximes have often been used as chelating ligands in the field of coordination chemistry and their metal complexes have been of great interest for many years.

The presence of mildly acidic hydroxyl groups and slightly basic nitrogen atoms makes oximes, dioximes amphoteric ligands, which form square planar, square-pyramidal or octahedral complexes with transition metal ions such as Co(III) and Ni(II) as the central atom (11). The structure of 1-2 naphthoquinone dioxime is examined by use of the HF (6 -31 G*level), density functional theory DFT (6 -31 G* level) & hybrid functional B3LYP. Using the optimized structure of the titled compound IR, NMR, and ultraviolet data is calculated and compared with experimental data. It shows good relation between theoretically calculated IR wave numbers & observed values for Mid – Far IR data (12). Al, Zn, Cu (II), Ni(II) and alkali metal salts of dioximes were reported and the authors concluded that the colour of the quinone oximes is not related to quinone oxime structure (13). In this paper we report synthesis of bivalent metal chelates of the type M [NQO]$_2$ where M = Hg, Cd, Zn, Pb: NQO = 1, 2 napthoquinone dioxime and Ag (NQO) and characterization by XRD, Mid IR, thermal analysis, electronic spectra, SEM photographs and antimicrobial activity against microorganisms have been studied.

II. MATERIALS AND METHODS

The ligand 1, 2-naphthoquinone dioxime is synthesized in laboratory as per the reported method (14). A stock solution of Pb (II), Zn (II), Cd (I), Hg (II) and Ag (I) is prepared by using AR grade chemicals. Deionised water is used during synthesis.

2.1 Preparation of metal chelates.

The chelates were prepared by mixing metal salt solution and ligand in 1: 1 proportion for silver chelate and 1: 2 for other metals. The mixture was constantly stirred for one hour on magnetic stirrer. The pH of the mixture was maintained, in between 5.0 – 6.0 by adding ammonia solution to it. Warm the mixture on water bath for about 15 minutes. On cooling it was filtered and found to be coloured.

2.2 Instrumental Analysis.

Elemental analysis was carried out with a Perkin Elmer 2400 series for C, H, O & N. The IR spectra are recorded on a Shimadzu FTIR 8400 S model in a KBr matrix. Electronic spectra were recorded on Perkin Elmer UV – visible spectrometer, Lambda-25 model. TGA curves were recorded on Perkin Elmer Pyris. TGA were recorded on apparatus using 10^{0}C/min in air and DSC on DSC 800 model. The proton and ^{13}C NMR spectra recorded in DMSO dueterated on Varion. 400 MR, powder x ray diffraction patterns were obtained by using Rigaku Mini flex (II). LC – MS scans were carried out on Shimadzu – LCMS 2010 employing electron impact source. Scanning electron microscopy was carried out on Vega 2SB model and EDAX on OXFORD INCA PENTA with TESCAN VEGA 2SB.

2.3 Antimicrobial activity testing

Test organisms: The antimicrobial activity of ligands, metal salts and synthesized metal chelates is tasted against bacteria [*Escherichia coli* (NCIM 2065), *Bacillus subtilis* (NCIM 2063), *Staphylococcus aureus* (NCIM 2079), *Proteus Vulgaris* (NCIM 2813), *P. aeruginosa* (NCIM 2200), *Aspergillius Niger* (NCIM 1196) and Candida *albicans* (NCIM 3471)] strains collected from NCL, Pune India.

2.4 Maintenance of culture:

The cultures of bacteria and fungi were maintained on Nutrient agent (Hymenia Laboratories Pvt Ltd. Ref. M 002-500G 99% Purity), Mueller-Hinton Agar (Himedia Laboratories Pvt. Ltd Ref. M 173 – 500G, 99% Purity) and subcultured accordingly and preserved at 4^{o} C. for 24 hours in incubator.

2.5 Plating

The 100 µL cell suspension (108 cell / ml of bacteria & yeasts C. albicans and 100 µL of spore suspension of mold (A niger) were spread on then. Agar (for bacteria) and Mueller-Hinton Agar for fungi were used. Then wells were bored in the media. In the wells DMSO (solvent), ligand, metal salts and metal chelates solutions were poured for each organism, and then incubated at 37^{0}C for 48 hrs. for bacteria and 30^{0}C for 5 days for fungi . The zone diameter of inhibition were measured in mm & recorded.

III. RESULTS AND DISCUSSION

3.1 Thermal Analysis:

The TG, and DTG data is given in Table 1. The ligand 1, 2 naphthoquinone dioxime (NQO) is heated with 10^{0}C/min in air up to 380^{0}C and DSC was carried out by heating 10^{0}C/min from 30^{0}C to 200^{0}C in N_2. The TG data of NQO shows first weight loss corresponds to the loss of [N-2(OH)] which is in good agreement in the range of 30 -245^{0}C (calculated weight loss is 48% and found as 47.89%). It indicates that the ligand has high thermal stability. DSC data of NQO shows one exotherm. The onset temperature at 143.25^{0}C and the peak temperature at 147.63^{0}C and ΔH found to be -219.50 J/g. It can be assigned to loss of [N-2(OH)] moiety.

TG data of metal chelates of Pb, Cd, Zn and Hg show more or equal results for the first step decomposition about 100 -350^{0}C and weight loss in range of 25 to 35 % and DTG temperatures are in the range of 152 ± 1^{0}C. Ag (NQO) shows weight loss in the temperature range 146 – 350 ^{0}C upto 22 % and DTG temperature is at 163^{0}C.

DSC data of metal chelates shows that the chelates containing Zn, Hg and Cd exhibits an exotherm starting at about 146^{0}C and peak temperatures about at 151 ± 2^{0}C. There is a variation of the absorption of heat as it can be seen from the Table 2. In the case of Ag (NQO) it shows an exotherm with onset temperature at 153.55^{0}C and peak temperature at 164.38^{0}C. The enthalpy of the process is recorded as -305.69J/g. Pb (NQO) 2 shows an exotherm which starts at 187.64^{0}C and peak temperature at 190.76^{0}C. The heat of decomposition is 130.80 J/g.

Table: 1 Thermo analytical data of ligand and its chelates

Sr.No.	Compound	Temp Range ^{0}C	% loss	DTG ^{0}C
1	NQO	122-245	48.0	150
2	Zn $(NQO)_2$	142-350	85.0	162
3	Cd $(NQO)_2$	100-350	47.5	152
4	Hg $(NQO)_2$	145-355	80.0	172
5	Ag(NQO)	100-350	53.0	164
6	Pb $(NQO)_2$	120-350	65.0	158

Table: 2 Differential Scanning Calorimetric data of ligand and its chelates

Sr.No.	Compound	DSC EXO Onset Temp. ^{0}C	Peak Temp. ^{0}C	ΔH J/g
1	NQO	143.25	147.63	-219.50
2	Zn $(NQO)_2$	148.40	154.5	-180.36
3	Cd $(NQO)_2$	130.65	123.67	-124.77
4	Hg $(NQO)_2$	146.20	150.75	-139.06
5	Ag(NQO)	145.90	159.50	-112.75
6	Pb $(NQO)_2$	134.69	143.87	-172.19

3.1.1 Kinetics

To calculate kinetic parameters of 1, 2-naphthoquinone dioxime a separate TG/DTG was run on Shimadzu STA in the ranges 0.0^{0}C to 800^{0}C in air. The decomposition started at 122.67^{0}C and ended at 223.98^{0}C. The observed weight loss was 48%. The TG data was used to calculate kinetic parameters by using developed software (15). The kinetic parameters for F-5 model calculated using Coates _ Redfern equation which is random nucleation (Mampel unimolecular law). The results are given in Table -3.

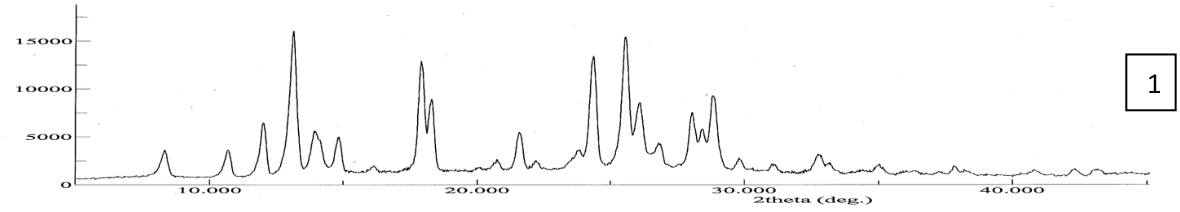
Table: 3 Kinetic parameters for decomposition of NQO in air

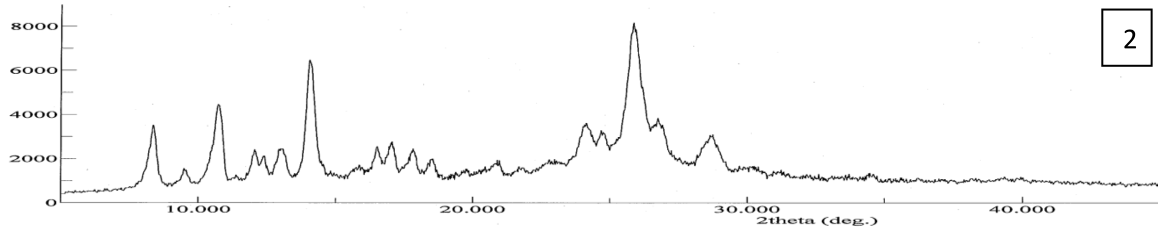
Sr. No.	Parameter	Step I 122.67 to 223.98 ^{0}C
1	n Order of reaction	1
2	E Energy of activation	44.12 KJ/Mol
3	R regression coefficient	0.848
4	Log A Frequency factor	2.4278
5	S Entropy of activation	-210.61 J/mol/K
6	G Free energy of activation	187.34 KJ/Mol
7	H Enthalpy of activation	91.9697 KJ/Mol

3.2 X-ray diffraction:

The x-ray diffraction data was recorded on Rigaku X-ray diffractometer (Cu K_α radiation) Miniflex 2. The data was processed by using McMaille computer program for determination of cell parameters and space group (16). 1, 2 naphthoquinone dioxime crystallizes in the triclinic group and it has crystallographic parameters,

a = 10.7684 A^o, b = 8.5280 A^o and c = 8.7887 A^o
$\alpha = 101.464^o$, $\beta = 102.764^o$, $\gamma = 85.881^o$,
Its volume is 771.106 $(A^o)^3$ and space group H-M symbol P1.
Dmin = 2.853032 g/cm^3





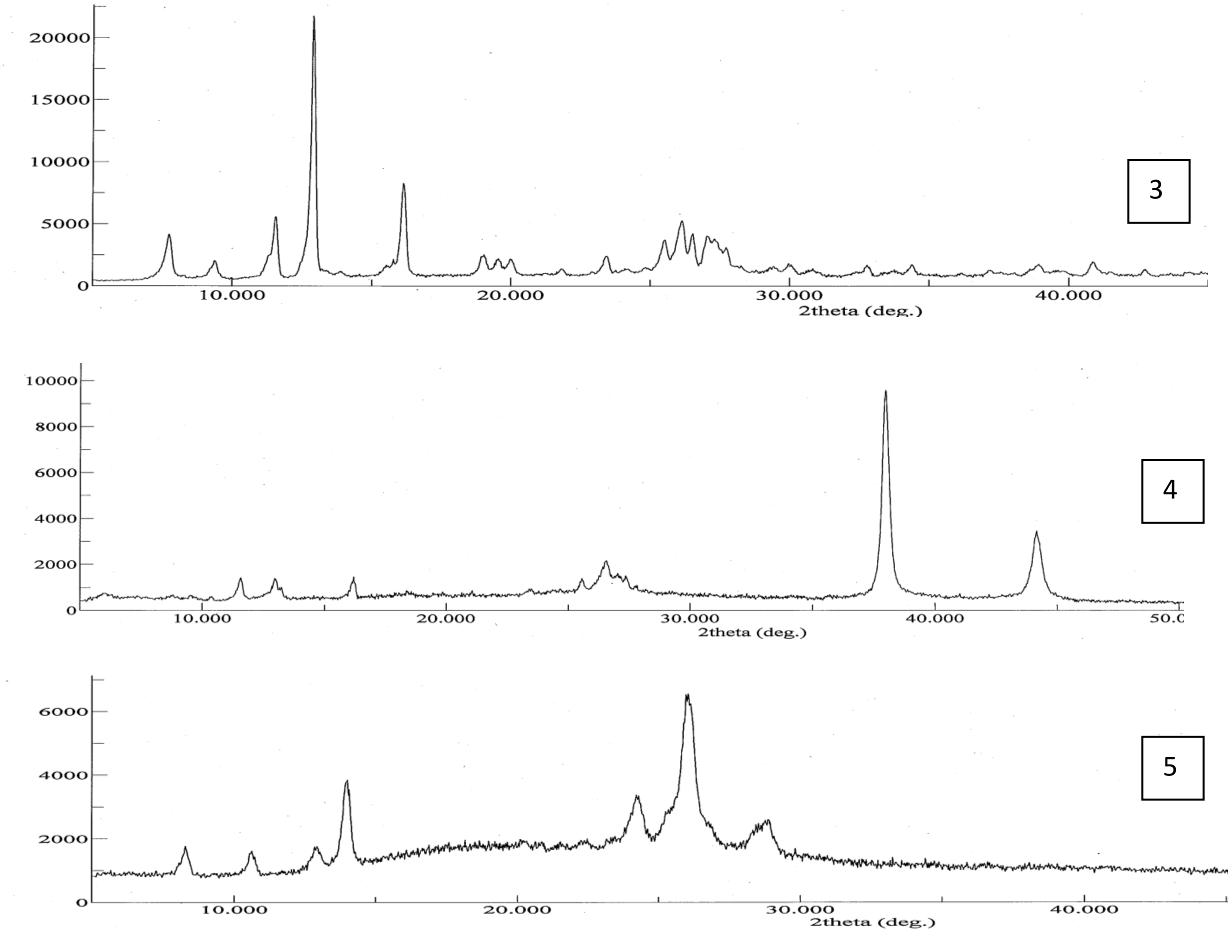


Fig.1 XRD PATTERNS OF METAL CHELATES 1) Pb $(NQO)_2$, 2) Hg $(NQO)_2$, 3) Zn $(NQO)_2$, 4) Ag (NQO) AND 5) Cd $(NQO)_2$

Table: 4 h k l values of NQO) and Th (Obs) and Th (Calc)

Sr.No.	h	k	l	Th(Obs)	Th-Zero	Th(Cals)	Diff.
1	1	0	0	8.440	8.834	8.415	0.020
2	0	1	0	10.580	10.574	10.580	-0.005
3	0	1	-1	11.980	11.974	11.994	-0.019
4	0	1	-1	13.440	13.434	13.442	-0.008
5	1	1	-1	14.460	14.454	14.464	-0.010
6	1	-1	-1	17.434	17.434	17.439	-0.004
7	0	0	2	21.100	21.096	21.084	0.008
8	1	2	0	22.680	22.674	22.658	0.016
9	2	0	-2	24.114	24.114	24.121	-0.007
10	2	2	-1	25.920	25.914	25.921	-0.006
11	2	2	0	26.860	26.654	26.861	-0.007
12	3	-1	0	29.820	27.914	27.903	0.011
13	3	-1	-1	28.640	28.634	28.636	-0.001
14	2	-1	-1	30.520	30.514	30.528	-0.014
15	3	-1	1	31.100	31.094	31.095	0.000

M. Nasakkala et.al. (17) have reported that NQO belongs to monoclinic, space group $P2_1/c$ (N.14), a = 7.082(12) A^0, b = 9.046(11) A^0, c = 13.845(11) A^0, β = 100.2 $(1)^0$, Z=4 and D=1.432 g/cm^3.

The metal chelate of Zn (NQO)2, shows data as per computer code referred above that it belongs to Triclinic, a = 16.9620, b = 9.7908 c = 7.0376 A^0, α = 104.005 °, β = 66.096 °, γ = 62.244 °, volume = 800.416 $(A^O)^3$ and density calculated as 3.388752 g/cm^3 with Z = 2. Table 5 shows h k l and theta values.

Table: 5 h k l values of Zn (NQO)$_{2,}$ Th (Obs) and Th (Calc)

Sr.No.	h	k	l	Th(Obs)	Th-Zero	Th(Cals)	Diff.
1	0	1	0	12.040	12.057	12.057	0.000
2	1	0	1	13.160	13.177	13.177	0.000
3	0	1	-1	14.140	14.157	14.172	-0.015
4	3	0	-1	17.940	170957	17.946	0.011
5	3	1	1	18.320	18.337	17.343	-0.005
6	1	2	-1	20.080	20.097	20.092	0.005
7	0	2	-1	20.760	20.777	20.769	0.008
8	1	0	-1	21.600	21.617	21.602	0.015
9	2	-1	0	24.400	24.417	24.430	-0.13
10	2	-1	2	25.620	25.637	25.640	-0.003
11	3	2	-1	26.120	26.137	26.141	-0.004

The metal chelate of Hg (NQO)2, shows data as per computer code referred above that it belongs to Triclinic, a = 6.7244, b = 9.3817 c = 12.0611 A^0, α = 84.957 ^{o}, β = 108.484 ^{o}, γ = 86.828 ^{o}, volume = 716.055 (A^O)^3 and density calculated as 3.388752 g/cm^3 with Z = 2. Table 6 shows h k l and theta values.

Table: 6 h k l values of Zn (NQO)$_{2,}$ Th (Obs) and Th (Calc)

Sr.No.	h	k	l	Th(Obs)	Th-Zero	Th(Cals)	Diff.
1	0	0	1	7.760	7.786	7.770	0.016
2	0	1	1	11.560	11.586	11.586	0.000
3	0	1	-1	12.920	12.946	12.934	0.012
4	1	0	0	13.880	13.906	13.928	-0.022
5	0	0	2	15.520	15.546	15.576	-0.030
6	1	1	0	16.160	16.186	160169	0.017
7	0	2	0	19.020	19.046	19.050	-0.004
8	0	0	3	23.440	23.466	23.456	0.010
9	1	1	2	24.820	24.846	24.846	0.000
10	1	1	-3	25.520	25.546	25.548	-0.002
11	1	2	-2	26.140	26.166	26.174	-0.008
12	2	0	-1	26.520	26.546	26.557	-0.001
13	1	-1	2	27.060	27.086	27.078	0.008
14	2	0	-2	27.340	27.366	27.350	0.16

The particle sizes were calculated using Debye Scherer Formula and the values found to be for Zn (NQO)$_2$ as 34.04 nm and for Cd (NQO)$_2$ as 19.23 nm which indicates that these crystals are nano crystals. Other chelates of Pb (NQO)$_2$, Hg (NQO)$_2$ and Ag (NQO) are crystalline in nature but their crystallographic analysis could not be done , only particle sizes were calculated and found to be 16.51 nm, 42.54 nm and 27.47 nm respectively.

3.3 Electronic Spectra (UV)

The UV spectra of the ligand NQO and its metal chelates M (NQO)$_2$ where (M = Zn, Cd. Hg Ag and Pb) were studied in a dimethyl sulphoxide (DMSO) solution and the data is complied in Table 7. NQO exhibits absorption bands at 211.2 nm and at 262.6 nm. These bands are assigned to π to π*. The band at 262.6 nm is originated from the π to π* of the orthoquinone oxime (7). The chelates, studied here show only single transition at 262.1 nm which is π to π* transition.

Table: 7 Electronic absorption data (λ nm) of metal chelates in DMSO in the range (200-800 nm).

Sr.No.	Compound	λ	
1	NQO	211.2	262.6
2	Zn (NQO)$_2$	201.5	263.7, 363
3	Cd (NQO)$_2$	202.8	260.7, 361.5
4	Hg (NQO)$_2$	202.4	267.4, 365.0
5	Ag(NQO)	202.1	267.3, 365.2
6	Pb (NQO)$_2$	201.3	266.3, 365.1

3.4 Infrared Spectra

IR frequencies of 1-2naphthoquinone dooxime were calculated by RHF / 6-31G* and reported by N.R. Gonewar et. al. (18). In IR spectra of chelates M $(NQO)_2$ where M = Pb, Zn, Hg and Cd showed a weak γ (C – H) stretching at about 3000 – 3400 cm^{-1}. The functional group such as C = O, C = N and N – O assigned. The data is given in table 8. It can be seen from the table that the spectrum of NQO can be compared with chelates of metals which clearly shows lower wave numbers for γ(C = N) and V (C = O) bands owing to elongation of these bonds upon coordination. The absorption of γ (N – O) was found at higher wave numbers since this bond was significantly shortened in the chelates. The high position of γ (NO) frequencies indicates that nitroso atom of the oxime group coordinates to the centre (19, 20).

Table: 8 Characteristic ν IR (cm^{-1}) bands of NQO and its metal chelates.

Sr.No.	Compound	C –H	C = O	C = N	N - O
1	NQO	3213	1608	1669	1093
2	Zn $(NQO)_2$	3271	1618	1666	1093
3	Cd $(NQO)_2$	3214	1591	1664	1062
4	Hg $(NQO)_2$	3282	1591	1658	1084
5	Ag(NQO)	3180	1589		1082
6	Pb $(NQO)_2$	3201	1568	1662	1084

3.5 Proton NMR and ^{13}C NMR

NQO shows chemicals shift of proton at 8.21 and 8.19 ppm for N-OH group.

Table: 9 Chemical shifts of metal chelates in ppm

Sr.No.	Compound	H1	H2
1	Zn $(NQO)_2$	12.69	11.94
2	Cd $(NQO)_2$	9.13	9.11
3	Hg $(NQO)_2$	8.67	8.65
4	Ag(NQO)	8.77	8.75
5	Pb $(NQO)_2$	8.67	8.65

The chemical shift for hydroxyl proton of metal chelates are shown in Table:9. It is observed that only Zn (NQO)2 shows higher chemical shifts as compared to other chelates. The remaining chemical shifts are similar to ligand molecule. (21, 22).

13C NMR chemical shifts of C_1 and C_2 where –NOH C_1 is bonded which are observed at 184.37 and at 181.22 ppm. The chemical shifts of the metal chelates are given in Table:10,

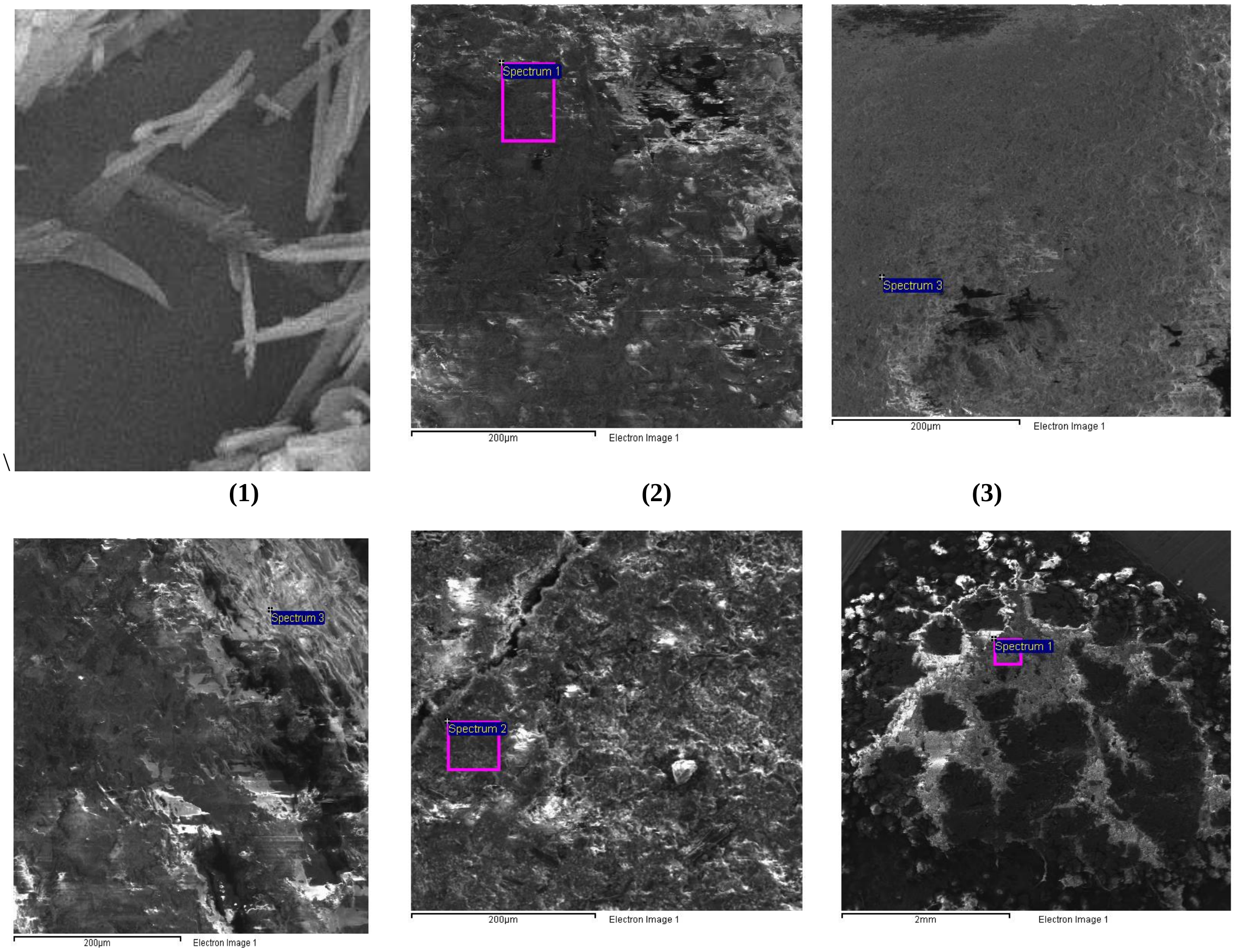
Table: 10 Chemical shifts of metal chelates in ppm

Sr.No.	Compound	C1	C2
1	Zn $(NQO)_2$	140.66	133.95
2	Cd $(NQO)_2$	147.35	140.81
3	Hg $(NQO)_2$	147.99	140.62
4	Ag(NQO)	140.62	133.94
5	Pb $(NQO)_2$	147.94	140.61

The chemical shift for C1 is observed in the range 140.62 to 147.99 ppm for the chelates under studies and that of C2 is observed in range 133.94 to 140.81 ppm. The chemical shifts of remaining carbons are in good agreement (23, 24).

3.6 SEM studies

The scanning electron microscopy (SEM) of the ligand and their Zn (II), Cd (II), Hg (II), Ag (I) and Pb (II) chelates was carried. In general, the average crystallite size of the metal chelates is smaller than the crystallite size of the parent ligand. These results of SEM investigations support the results obtained from XRD investigations. The XRD patterns were found to be composed of overlapped sharp lines as well as broad bands indicative of both small crystalline of nano crystalline type and extremely small crystallite size tending to amorphous nature. Secondly a careful examination of the SEM photographs (shown in Fig.2) of the ligand and their five metal chelates reveals that all the samples are heterogeneous mixtures of different particle size.



(1) **(2)** **(3)**

(4) **(5)** **(6)**

Fig.2 SEM DATA OF 1) NQO, 2) Zn(NQO)$_2$, 3) Cd(NQO)$_2$, 4) Hg(NQO)$_2$, 5) Ag(NQO) and 6) Pb(NQO)$_2$

3.7 Antimicrobial Scanning Results

The antimicrobial activity of metal salts, ligands and their complexes were tested against bacteria and fungi like *Escherichia coli*, *Bacillus subtilis*, *Staphylococcus aureus*, *Proteus vulgaries*, *Pseudomonas aeruginosa*, ESBL *Escherichia coli*, *Klebsiella Pneumoniae*, *Aspergillus niger*, *Candida albicans.*The causative agent Cisplatin is chosen as standard chemotherapy agent.

The testing against growth of micro-organisms was carried out by using well diffusion method employing Mueller Hinton Agar (MHA) and culture in nutrient broth in each case of micro-organisms. The concentration of NQO and its metal chelates were chosen as 10^{-4}M. The plates were incubated at 35^{0}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: 11 Dimethyl sulphoxide i.e. solvent used shows no inhibition for all organisms under studies.

Table 11: Antimicrobial activities of 1, 2 naphthoquinone dioxime (NQO) and its metal chelates (Inhibition zone diameter in mm)

Compound	***E. coli***	***B. subtilis***	***S. aureus***	***Proteus vulgaries***	***P. aeruginosa***	**ESBL *E. coli***	***K. Pneumoniae***	***A. nigar***	***C. albicans***
DMSO	Nil	Nil	Nil	Nil	Nil	Nil	Nil	Nil	Nil
NQO	20 s	24 b	16 s	23 b	10 w	18 s	18 s	11 w	13 w
$Zn\,(NQO)_2$	22 b	19 s	28 Sh	17 s	24 b	14 w	23 b	19 s	20s
$Cd(NQO)_2$	18 s	20 s	25 b	25 b	23 b	14 w	15 w	16 s	22 b
$Hg(NQO)_2$	17 s	20 s	23 b	15 w	26 S	13 w	14 w	17 s	19 s
Ag(NQO)	17 s	25 b	24 b	22 b	20 s	13 w	14 w	18 s	19 s
$Pb\,(NQO)_2$	19 s	28 Sh	30 Sh	23 b	13w	20 s	17 s	18 s	21 b
Cisplatin	18 s	13 w	20 s	18	No test	No test	No test	No test	No zone

(10-15) Weak; (16-20) Small; (21-25) Broad; (26-above) Sharp zone

Metal complexes and ligands by Agar well diffusion method showing significant growth inhibition against all the microorganisms. At 5 mg/ml of concentration, all the chemicals shows intermediary zone of inhibition (11 to 31 Mm). The inhibition effects of metal complexes on the microbial growth are significantly different than metal salts. Table No. 4.4.2 shows that, metal salts such as Zn, Ag and Pb salts shows weak inhibition zones than others. It revealed that, these metal salts have less and their complexes have moderate antimicrobial effect.

The species *P. aeruginosa*, *A. niger*, *C. albicans* shows nearly similar range of zone diameter (13-27mm, 16-28mm, 12-30mm resp…) at 7mg/ml of salt concentration; which was shown by the species *E. coil*, *B. subtilis, S. aureus*, *P. valgaries* at 5mg/ml of salt concentration.

The antibacterial potency increases upon complexation as compared to uncomplexed one (Metal salts). So the biological activity of metal salts has been enhanced with the addition of ligands. It is suggested that positive charge of metal ion is partially shared with the donor atoms and there is electron delocalization over the whole chelate ring system having center anion, which effectively influence the solubility mechanism of the complex. It is however indicates, that chelation tends to make the metal salts act as more powerful and potent bactericidal agents, thus killing more of the microorganisms than the parent metal salts.

It is suspected that factors, such as solubility, conductivity, and dipole moment and cell permeability mechanisms (influenced by the presence of metal ions) may be possible reasons for increasing this biological activity.

The result also indicates that the metal complexes (Chelates) are found to be potentially more active against bacteria than the fungi. $Pb\,(NQO)_2$, Ag (NQO), $Hg\,(NQO)_2$ and $Cd\,(NQO)_2$ showed highest activity for P. vulgaris. The inhibition of the micro-organism growth for metal chelates was found to be in the following order.

Zn = Hg > Cd > Ag = Pb > Cisplatin

Cisplatin was the first member of a class of platinum-containing anti-cancer drugs. **Cisplatin** is particularly effective against testicular cancer. The studies demonstrate that metal chelation can increase the anti microbial activity than metal free ligand. It is reported that metal chelation reduces the polarity of the metal ion mainly due to the 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 (25). It favours 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 increases the electrostatic filed around the metal ion.

IV. CONCLUSIONS

Most of the synthesized metal complexes have shown biological activities against the tested nine types of both bacterial and fungal species by Agar well diffusion method. To counter the increased bacterial, fungal, viral, and microbial, cancer and aids, etc. diseases to the rapid growth in population, investigation of the biological activities of the ligands and their metal complexes would be very interesting and useful to control the respective diseases. These results are better than standard Cisplatin chemotherapy agent. All these chelates are crystalline in nature and generally belong to triclinic. The coordination ability of NQO towards M (II) chelates were examined by different spectroscopic methods that unequivocally determine the coordination sites of NQO. It is observed that the ratio of metal to NQO is 1:2 for chelates of Zn, Pb, Cd and Hg while in case of Ag the ratio is 1:1.

ACKNOWLEDGEMENT

We thank Prin. K.D. Jadhav, Principal, Bharati Vidyapeeth Deemed University, Yashwantrao Mohite College, Pune for permission to Publish this work.

REFERENCES

[1]. V. B. Jadhav, N.R. Gonewar, K.D. Jadhav and R.G. Sarawadekar, Investigation on theoretical calculations of mid-far infrared, NMR and electronic spectra of 1-nitroso-2, naphthol or 1-2 naphthoquinine-1, oxime and comparison with experimental data, Journal of Pharmacy Research 2011,4(12),

[2]. N.R. Gonewar, V. B. Jadhav K.D. Jadhav, and R.G. Sarawadekar, Theoretical calculations of infrared, NMR and electronic spectra of 2-nitroso-1, naphthol or 1-2 naphthoquinine-2 oxime and comparison with experimental data, Research in Pharmacy 2(1) : 18-25, 2012.

[3]. Feigl F., Chemistry of Specific, Selective and Sensitive Reactions, Academic, New York, 1946.

[4]. Cukurovali A, Tas E, Ahmedzad M, Synthesis and Metal Complexation of a New Oxime

[5]. Derivative of 2-Aminothiazole, Synth React Inorg Met Org Nano Met Chem, 27(5),

[6]. 1997, 639– 646.

[7]. Mukherjee S, Patel B A, Bhaduri S, Selective Ethylene Oligomerization with Nickel Oxime Complexes, *Organometallics*, 28 (10), 2009, 3074–3078.

[8]. Ponomareva VV, Dalley NK, Xiaolan K, Gerasimchuk NN, Domasevich KV, Synthesis, spectral and crystal structures of complexes of ambidentate C6H5C(O)C(NO)CN– ,J Chem Soc Dalton Trans, 1996, 2351-2359.

[9]. Milios CJ, Stamatatos TC, Perlepes S P, The coordination chemistry of pyridyl oximes, Polyhedron, 25, 2006, 134-194.

[10]. Alzoubi B m, Vidali F, Puchta R, Dücker-Benfer C, Felluga A, Randaccio L, Tauzher G, Van Eldik R., Mechanistic behaviour of alkylcobaloximes and imino-oxime complexes related to vitamin B(12), Dalton Trans, 7(13), 2009, 2392-2399.

[11]. Alper T O, Gazi I, Hasene M, Tuncer H, Nagihan A, A Co(III) complex with a tridentate amineimine- oxime ligand from 1,2,3,4-tetrahydroquinazoline: synthesis, crystal structure, spectroscopic and thermal characterization , J Coord Chem, 62(6), 2009, 1005–1014.

[12]. Kufelnicki A, Fritsky IO, Sliva TY, Golovaneva IF, Lampeka R D, Stereoselective synthesis of cobalt(III) anionic complexes with chiral pyruvylaminoacid oximes and metal–ligand interactions in aqueous solution, Polyhedron, 26(12), 2007, 2894-2900.

[13]. Gok Y, Kantekin H**,** The synthesis and characterization of new (*E*,*E*)-dioxime and its mono andconheteronuclear complexes containing 14-membered tetraaza macrocyclic moiety, Polyhedron, 16(14), 1997, 2413-2420.

[14]. V.B. Jadhav, N.R. Gonewar, K.D. Jadhav and R.G. Sarawadekar, Synthesis, spectral and Theoretical investigation on 1-2 naphthoquinone dioxime, Research in Pharmacy 1(3) : 01-09, 2011.

[15]. A.P. Avdeenko, G.M. Glinyanaya and V.V. Pirozhenko, Russian Journal of Organic Chemistry, 35(10)1480-1487 (1999).

[16]. M.N. Huges, Inorganic Chemistry of Biological Processes. Willy, New York (1981).

[17]. R.G. Sarawadekar and I.B. Gade, Software for Kinetic Analysis of non isothermal thermogravimetry (TG) Data, HEMRL REPORT , PYRO, TR 253 (2006).

[18]. A. Le Bail, Powder Diffraction, 19, 249-259 (2004).

[19]. M. Nasakkala, H. Saarinen, J. Korvenrata and E. Nasakkala, The Crystal Structure of (1Z, 2E) – 1-2 Naphthoquinone Dioxime, Acta. Chemica Scandinavica, A 31 469 – 472 (1977).

[20]. V.B. Jadhav, N.R. Gonewar, K.D. Jadhav and R.G. Sarawadekar, Synthesis, spectral and Theoretical investigation on 1-2 naphthoquinone dioxime, Research in Pharmacy 1(3) : 01-09, 2011.

[21]. Constantino V. R. Toma H. E., Olivenia LFC and P. S, Snatos, *J. Raman Sepctroscopy*, 23, 629 (1992).

[22]. Natarajan C., Nazeer Hussain A. *Indian J. Chem*. 22A, 527 (1983).

[23]. A. Krzan and J. Mavri., *Chemical Physics*, 277, 71-76 (2002).

[24]. V. Enchev, G. Ivanova, A. Vgrinov, G. D. Neykov., *J. Mol, Struct.* 508, 149 (1999).

[25]. G. Ivanov, V. Enchev. *Chem.. Phys*. 264, 235 (2001).

[26]. T. Shono, Y. Hayashi and K. Shinra., *Bull. Chem. Soc. of Japan*., 4, 3179-3181 (1971).

[27]. Z.H. Chohan, C. T. Superan and A. Scozafava, *J. Enzyme Inhihitian and Medicinal Chemistry*, 20, 303 (2005).