

SYNTHESIS, CHARACTERIZATION AND ANTIMICROBIAL STUDIES OF CO-ORDINATION POLYMERS

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(Received: May 14, 2012 - Accepted: August 17, 2012)

ABSTRACT

A novel heteronuclear bis-ligand namely 5-[4-(2-carboxyphenyl carbonyl amino)-phenyl-4-carbonyl methyl amino]-8-hydroxyquinoline was synthesized and characterized. This Bis-ligand was designated as CPHQ. Co-ordination polymers of this CPHQ bis-ligand were prepared with Cu⁺², Co⁺², Ni⁺², Mn⁺² and Zn⁺² metal ions. All of these Co-ordination polymers and the CPHQ ligand were characterized by elemental analysis, IR, NMR spectral studies, thermogravimetry, electronic reflectance spectral studies and magnetic susceptibility measurement. The synthesized novel Bis-ligand and their Co-ordination polymers were screened for their antimicrobial activity.

Key Words: Heteronuclear Bis-ligand; Co-ordination polymer; spectral studies; magnetic moment; antibacterial and antifungal activities.

INTRODUCTION

The research on Co-ordination polymers by linking transition metal ions with ligands has been constantly developed in past years. Because of their excellent properties, such as semiconducting catalytic properties, waste water treatment for metal recovery, in protective coating, as antifouling paints and anti fungal properties^{1,2}. Recently new coordination supramolecules and polymers bis(oxine), bidentate ligand based on transition metal compounds and multidentate organic ligand has attracted much interest. In this context bis(oxine) ligand with two oxine units link by a bridge of 5,5'-methylene (-CH₂-), 5,5'-sulfonyl (-SO₂-), 5,5'-dimethylene sulfide (-CH₂-S-CH₂-) and -CH₂-O-CH₂- are reported in literature³⁻⁹. Most of bichelating ligands are derived from well known chelating agents like 8-hydroxy quinoline and salicylic acid etc^{3,6}. 8-Hydroxyquinoline (8-HQ) moiety has received constant consideration due to their efficient ionophores, therapeutic and fluorescences properties¹⁰⁻¹⁴. 8-Hydroxyquinoline containing polymers are used in areas such as waste water treatment to recover metals, protective coatings, water disinfectants, ion-exchange resin, antifouling paints, antimicrobial, surgical materials, gels and ointment for medical uses¹⁵⁻²¹. 8-Hydroxyquinoline containing polymers and copolymers reveal complexing ability and biological activity²²⁻²⁵. The study of 8-quinolinol containing polyester has also been reported²⁶. The area in which the Co-ordination polymers having bis-azo dye containing ligands has not been developed so far. Such ligand may afford the Co-ordination polymer with different properties. Hence, it was thought interesting to explore the field of Co-ordination polymers based on heteronuclear Bis-ligand having 8-hydroxyquinoline and amic acid. Though the amic acid is generally derived by condensation of amines with anhydrides having carboxylic and amide group. The metal complexation study of various amic acids has been reported recently²⁷⁻³⁰, so the proposed present work is in connecting with the Co-ordination polymers based on heteronuclear Bis-ligand. The synthetic route for the preparation of bis-ligand and its Co-ordination polymers are shown in scheme 1.

EXPERIMENTAL

Materials

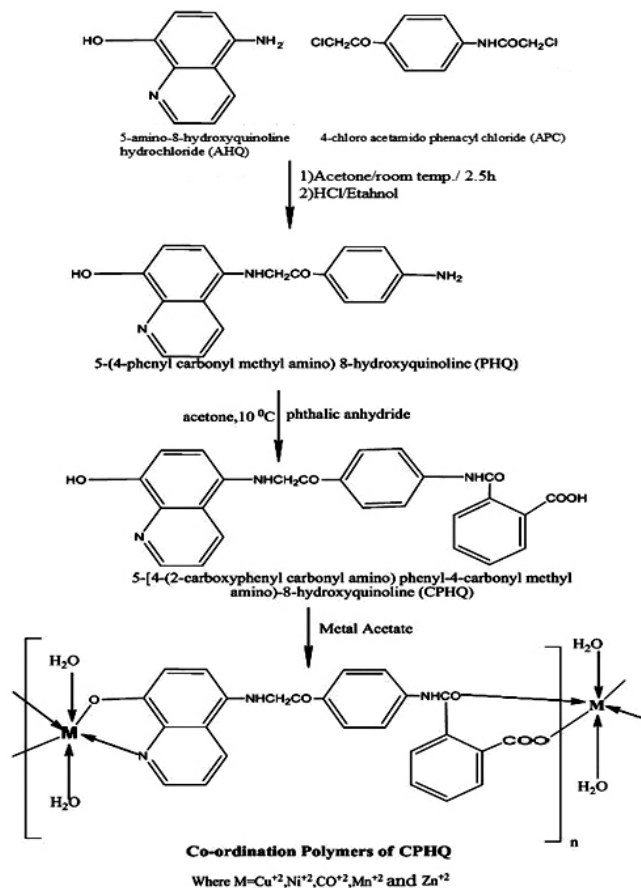
All the chemicals used were of analytical grade. The compound 5-amino-8-hydroxyquinoline hydrochloride (AHQ) was prepared by reported methods.^{31,32} 4-chloro acetamido phenacyl chloride (APC) was prepared by known method.³³

Synthesis of 5-[4-(2-carboxyphenyl carbonyl amino)-phenyl-4-carbonyl methyl amino]-8-hydroxyquinoline (CPHQ): The 5-[4-(2-carboxyphenyl carbonyl amino)-phenyl-4-carbonyl methyl amino]-8-hydroxyquinoline CPHQ was prepared in two steps.

1. Synthesis of 5-(4-phenyl carbonyl methyl amino)-8-hydroxyquinoline (PHQ): A solution of 4-chloro acetamido phenacyl chloride (APC) (24.6 g, 0.1 mole) in acetone was treated with 5-amino-8-hydroxyquinoline hydrochloride (AHQ) (23.1 g, 0.1 mole) in acetone at room temperature for 2.5 hours. The resultant product was filtered and hydrolyzed by 50:50 HCl and EtOH mixture.

The yield of CPHQ was 82 % (20.17 g); M.p.139-142 ° C (uncorrected).

2. Synthesis of 5-[4-(2-carboxyphenyl carbonyl amino)-phenyl-4-carbonyl methyl amino]-8-hydroxyquinoline (CPHQ): A solution of 5-(4-phenyl carbonyl methyl amino)-8-hydroxyquinoline (PHQ) (36.9 g, 0.1 mole) in acetone was cooled to 10 ° C and then a solution of phthalic anhydride (29.6 g, 0.2 mole) was added with constant stirring. The resulting product filtered and air-dried. The yield of CPHQ was 67 % (24.72 g); M.p. 194-196 ° C (uncorrected). Anal. Found for C₂₅H₁₉N₃O₅ (441): C, 67.9; H, 4.2; N, 9.4. Calcd. C, 68.02; H, 4.30; N, 9.52. (Scheme-1)



Scheme 1

PREPARATION OF CO-ORDINATION POLYMERS

All Co-ordination polymers were synthesized by using metal acetates in a general method described as follows:

A warm and clear solution of CPHQ (4.41 g, 0.01 mole) in 20 % aqueous formic acid (200 mL) was added to a solution of copper acetate (1.99 g, 0.01 mole) in 50 % aqueous formic acid (50 mL) with constant stirring. After complete addition of metal salt solution, the pH of reaction mixture was adjusted to about 5 with dilute ammonia solution. Polymeric chelates were separated out in the form of suspension and then digested on a water bath for one hour and eventually filtered, washed with hot water followed by acetone and N,N-dimethyl formamide (DMF) and then dried in air at room temperature. The yields of all Co-ordination polymers were almost quantitative. The obtained solid metal complexes and their colors are shown in Table 1. The complexes are stable solids, decomposing above 258 ° C without melting and insoluble in diethyl ether, acetone, ethanol, methanol and chloroform; however, they are soluble in dimethyl sulfoxide (DMSO) and DMF.

MEASUREMENTS

Elemental analysis of CPHQ and its Co-ordination polymers were carried out on a C, H, N elemental analyzer (Italy). IR spectra of the Bis-ligand and the polymeric chelates were scanned on a Nicolet-760D FTIR spectrophotometer in KBr. ¹H and ¹³C NMR spectra were carried out at room temperature in DMSO-d₆ and tetramethylsilane (TMS) as an internal reference using Bruker spectrophotometers (400 MHz and 100 MHz).

The metal content analyses of the polymeric chelates were performed by decomposing a weighed amount of each polymeric chelates followed by EDTA (disodium ethylene diamine tetra acetate) titration as reported in the literature.⁸

Magnetic susceptibility measurements of all the polymeric chelates were carried out at room temperature by the Gouy method. Mercury tetrathiocyanatocobaltate (II) was used as a calibration agent. Molar Susceptibilities were corrected for diamagnetism of component atoms using Pascal's constant. The diffuse reflectance spectra of the solid polymeric chelates were recorded on a Beckman DK-2A spectrophotometer with a solid reflectance attachment. MgO was employed as the reference compound. The number average molecular weight (\bar{M}_n) of all the Co-ordination polymers were determined by method reported in earlier communications.³⁴ Thermogravimetric analysis of Co-ordination polymers were carried on DuPont 950 TGA analyzer in air at a heating rate of 20 ° C/min.

Screening of antibacterial and antifungal activities

Antibacterial activities

In vitro antibacterial screening of CPHQ and its Co-ordination polymers were studied against gram-positive bacteria (*Bacillus subtilis* and *Staphylococcus aureus*) and gram-negative bacteria (*Escherichia coli* and *Salmonella typhi*) by cup-plate method³⁵ using nutrient agar as medium. In a typical procedure, molten nutrient agar kept at 45 ° C was then poured into Petri dishes and allowed to solidify. Then holes of 5 millimeter diameter were punched carefully using a sterile cork borer and these were completely filled with test solutions (1 mg/mL in DMF). The diameter of the zone of inhibition for all the compounds was measured and the results were compared with the standard drug streptomycin of the same concentration as that of the test compound under identical conditions.³⁶

TABLE 1. Analytical and spectral data of the CPHQ and its Co-ordination polymers.

Ligand/Co-ordination polymers (Empirical Formula)	Yield (%)	Color	Molecular Weight	Analyses Found (Calcd.) %				m_{eff} (B.M.) ^a	$(\bar{M}_n)_{60}^{\pm}$	\bar{D}_p ^c
				M	C	H	N			
CPHQ C ₂₅ H ₁₉ N ₃ O ₅	67	Light yellow	441	-	67.9 (68.02)	4.2 (4.30)	9.4 (9.52)	-	-	-
[Cu(CPHQ)(H ₂ O) ₂] _n Cu.C ₂₅ H ₁₇ N ₃ O ₅ .2H ₂ O	78	Light Green	538.54	11.6 (11.79)	55.5 (55.70)	3.6 (3.89)	7.6 (7.79)	1.92	3280	6
[Co(CPHQ)(H ₂ O) ₂] _n Co.C ₂₅ H ₁₇ N ₃ O ₅ .2H ₂ O	74	Brown	533.94	10.8 (11.03)	56.0 (56.18)	3.7 (3.93)	7.7 (7.86)	4.85	2716	5
[Ni(CPHQ)(H ₂ O) ₂] _n Ni.C ₂₅ H ₁₇ N ₃ O ₅ .2H ₂ O	80	Green	533.71	10.8 (10.99)	56.1 (56.21)	3.7 (3.93)	7.7 (7.86)	3.20	3248	6
[Mn(CPHQ)(H ₂ O) ₂] _n Mn.C ₂₅ H ₁₇ N ₃ O ₅ .2H ₂ O	76	Light Green	529.94	10.2 (10.36)	56.4 (56.61)	3.8 (3.96)	7.8 (7.92)	5.44	2698	5
[Zn(CPHQ)(H ₂ O) ₂] _n Zn.C ₂₅ H ₁₇ N ₃ O ₅ .2H ₂ O	82	Yellow	540.38	11.9 (12.01)	55.4 (55.51)	3.7 (3.88)	7.6 (7.77)	Diamagnetic ^d	2754	5

^a Magnetic susceptibility; ^b Number average molecular weight; ^c Average degree polymerization; ^d Diamagnetic behaviour.

Antifungal activities

The antifungicidal activity of all the compounds was evaluated against *penicillium expansum*, *Nigrospora Sp.*, *Trichothesium Sp.*, and *Rhizopus nigricum* by cup-plate method cultured on potato-dextrose agar medium adapting similar procedure describe above. The plates were incubated at 37 ° C for 48 hours. The diameter of the zone of inhibition for all the compounds was measured and the results were compared with standard drug Chlotrimazole in the same experimental conditions.³⁷

RESULTS AND DISCUSSION

The synthesis of the Bis-ligand, 5-[4-(2-carboxyphenyl carbonyl amino) phenyl carbonyl methyl amino]-8-hydroxyquinoline (CPHQ) has not been reported in the literature. All the metal complexes were sparingly soluble in common organic solvents but soluble in DMF, DMSO and Acetonitrile. The results of elemental analyses of Bis-ligand CPHQ and its Co-ordination polymers are agreed with those predicted on the molecular formula (Table 1).

The analytical data indicates that the metal complexes are agreed well with 1:1 metal to ligand (M/L) stoichiometry. The structure of CPHQ and its Co-ordination polymers was confirmed by the elemental analysis, FTIR, ¹H-NMR and ¹³C-NMR, reflectance spectra, magnetic susceptibility measurements and TGA analysis. Elemental (CHN) analysis, physical properties and IR data provided good evidence that the chelates are polymeric in nature.^{38,39} Possible structure of the Co-ordination polymers is shown in scheme 1.

Magnetic susceptibility

The magnetic susceptibility measurements of the metal complexes were performed at room temperature (Table 1). The magnetic susceptibility value for Cu²⁺ complexes of the ligand CPHQ is 1.92 B.M., it is less than the normal value⁴⁰ (1.84-2.20 B.M.). The lowered magnetic moment value observed for Cu²⁺ complex under the present study is due to distorted octahedral geometry.^{41,42} On the other hand magnetic moment values of Co²⁺ and Ni²⁺ complexes indicate an octahedral geometry for these complexes.⁴³ Mn²⁺ complex exhibited magnetic moment at 5.44 B.M.⁴⁴ It indicates that the complex is high-spin type paramagnetic, it lies within the octahedral range. The obtained result is very close to spin value 5.90 B.M. as the ground term is ⁶A_{1g} and thus supports the octahedral geometry.⁴⁵

Infrared spectra

The IR spectrum of CPHQ shows a broad band at 3340 cm⁻¹ attributed to the -OH stretching vibrations. The weak bands around 2862 and 2945 cm⁻¹ may be due to asymmetric and symmetric stretching vibrations of methylene groups. The bands at 3400 cm⁻¹ and 1710 cm⁻¹ are due to NH (Sec) and CO group respectively. The bands around 1634, 1575, 1500 and 1440 cm⁻¹ are attributed to the 8-hydroxyquinoline nucleus.¹⁸ The band at 1690 cm⁻¹ is recognized COOH group. The bands around 1650, 1610 and 1520 cm⁻¹ are attributed to amide group. Comparison of the IR spectrum of the Bis-ligand CPHQ and those of the Co-ordination polymers reveals certain characteristic differences. The broad band at 3340 cm⁻¹ for CPHQ has almost disappeared for the spectra of polymers. However, the weak band around 3200 cm⁻¹ in the spectra of CPHQ-Co²⁺, CPHQ-Ni²⁺, CPHQ-Mn²⁺ indicate the presence of water molecules which may have been strongly absorbed by the Co-ordination polymers. The C-O stretching in the ligand may be suggested due to the absorptions at 1366 and 1252 cm⁻¹. In the complexes, these bands are shifted to 1347 and 1245 cm⁻¹ respectively. These results indicate the formation of C-O-M bond. The peak at 1234 cm⁻¹ in the ligand is assigned to OH-bending of the phenolic moiety. This band is absent in the Co-ordination polymers. In addition to these bands, the spectrum of CPHQ has many characteristic absorption bands, which are identical to those that occur in 5-(4-phenyl carbonyl methyl amino)-8-hydroxy

quinoline (PHQ) and phthalic anhydride. The new bands observed in the region 470-440 cm⁻¹ and 430 cm⁻¹ are probably due to the formation of M-O and M-N bonds respectively.⁴⁶ Thus, it is possible that the oxygen and nitrogen atoms of 8-hydroxyquinoline groups are coordinated to the metal.^{47,48} From the above data the following structure has been proposed for the Co-ordination polymers shown in scheme 1.

¹H NMR and ¹³C NMR spectra of the Bis-ligand (CPHQ)

¹H NMR spectrum shows signal at δ 4.8 ppm (s, 2H) which is due to -CH₂ protons. Also single at δ 4.6, 9.4 ppm (s, 2H) due to -NH proton and single at δ 5.7 ppm (s, 1H) due to -OH proton. The signal at δ 8.9-6.9 ppm (m, 13H) is assign to aromatic protons of quinolone and phenyl moieties. A sharp peak at δ 11.3 ppm (s, 1H) is assign to -COOH group. These all features confirm the proposed structure of CPHQ Bis-ligand.

In the ¹³C NMR spectrum of CPHQ is observe a signal at 60.2 ppm it is assign to methylene carbons. Signals at 164.6-185.8 ppm are assign to C=O carbons. The signals observe between 112.5-150.6 ppm are assigned to aromatic carbons of the ligand.

Electronic spectra

The diffusion electronic spectrum of Cu²⁺, Co²⁺, Ni²⁺, Mn²⁺ and Zn²⁺ complexes were recorded in DMF. The diffusion electronic spectrum of CPHQ-Cu²⁺ Co-ordination polymers shows two broad bands at 15,380 cm⁻¹ and 22,730 cm⁻¹. The first bands may be due to ²T_{2g} → ²E_g transition, while the second may be due to charge transfer. The first band assigns distorted octahedral structure for the CPHQ-Co²⁺ complex. The higher value of μ_{eff} of the CPHQ-Cu²⁺ polymer support distorted octahedral structure.^{49,50} The CPHQ-Ni²⁺ and CPHQ-Co²⁺ polymers give two absorption bands at 17,250 and 24,000 cm⁻¹ and at 17,240 and 23730 cm⁻¹ respectively. These bands can be assigned to ⁴T_{1g} → ²T_{2g} and ⁴T_{1g} → ⁴T_{1g(P)} transitions respectively. The absorption bands and the values of μ_{eff} indicate an octahedral configuration for the CPHQ-Ni²⁺ and CPHQ-Co²⁺ polymers.^{51,52} The spectrum of CPHQ-Mn²⁺ showed weak bands at 16,474, 17,690 and 23,164 cm⁻¹ assigned to the transitions ⁶A_{1g} → ⁴T_{1g} (4G), ⁶A_{1g} → ⁴T_{2g} (4G) and ⁶A_{1g} → ⁴A_{1g} (4E_g) respectively, suggesting an octahedral structure for the Mn²⁺ complex.⁵² The spectrum of the Zn²⁺ complex is not well resolved, so it is not interpreted, but its μ_{eff} value shows that it is diamagnetic as expected.

Thermogravimetric Analyses

The thermal behavior of Co-ordination polymers and parent bis-ligand was investigated by TGA analysis. The TGA data of all the samples are presented in Table 2. The weight loss of the polymer samples at different temperature indicates that the degradation of the polymers is noticeable beyond 250 °C.

Table 2. Thermogravimetric analysis of Bis-ligand (CPHQ) and their Co-ordination polymers.

Ligand/ Co-ordination polymers	% Weight loss at different temperature (° C)							Activation energy (Kcal/mol)
	100	200	300	400	500	600	700	
CPHQ	1.4	6.8	40.4	56.9	60.4	63.1	66.0	9.7
[CuCPHQ (H ₂ O) ₂] _{n=2}	1.3	8.1	11.6	26.4	30.5	33.5	37.2	6.6
[CoCPHQ (H ₂ O) ₂] _{n=2}	5.7	6.9	12.8	26.3	38.0	43.2	49.9	8.6
[NiCPHQ (H ₂ O) ₂] _{n=2}	4.6	8.6	13.7	24.5	39.2	42.3	49.4	8.7
[MnCPHQ (H ₂ O) ₂] _{n=2}	3.8	6.3	12.1	22.6	31.4	34.6	36.2	8.4
[ZnCPHQ (H ₂ O) ₂] _{n=2}	4.1	7.5	17.6	33.9	41.8	49.1	52.3	9.5

A very slight decrease in weight loss depicted from the thermogram in the temperature range 50-150 °C for the parent ligand may be attributed to loosely bonded moisture. However, the initial slow weight loss occurring below 150 °C in all of the Co-ordination polymers may be attributed to the removal of water.

Inspection of the thermograms of all coordinated polymer samples reveals appreciable weight loss in the range 150 to 270 °C which might be due to metal-Co-ordinate water molecules (Scheme 1).³⁴ It was reported that the water molecules are coordinated to the metal ions.⁵³ The rate of degradation becomes maximum at temperature between 400 °C and 500 °C. Above this temperature, Co-ordination polymers are most stable and each Co-ordination polymers loses about 55 % of its weight when it is heated at 700 °C originating metal oxides.⁴⁷

The thermodynamic activation parameter of the decomposition process

of the Co-ordination polymers such as energy of activation (E_a) and order of reaction (n), were evaluated graphically by employing the Freeman-Carroll method⁵³ using the following relation:

$$[(-E_a/2.303R)\Delta(1/T)]\Delta\log W_r = -n + \Delta\log (dW/dt) \Delta\log W_r$$

where T is the temperature in K, R is gas constant, W_r = W_c-W; W_c is the weight loss at the completion of the reaction and W is the total mass loss up to time t. E_a and n are the energy of activation and order of reaction respectively. A typical curve of [Δlog(dw/dt)/Δlog W_r] vs [Δ(1/T)/Δlog W_r] for the Co²⁺ Co-ordination polymer is shown in Figure 1. The slope of the plot gave the value of E_a/2.303R and the order of reaction was determined from the intercept.

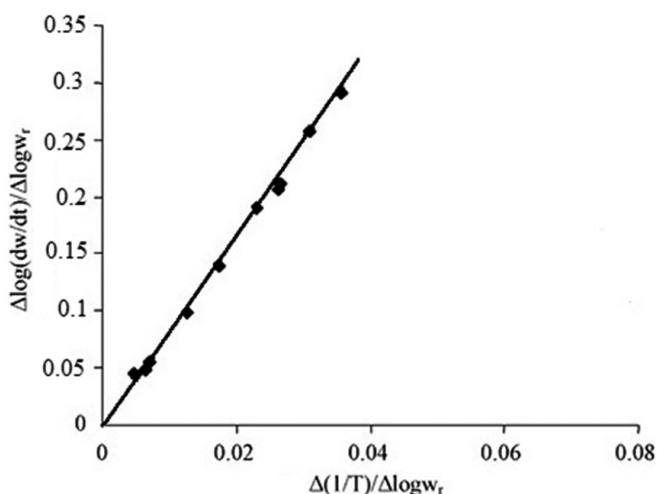


Figure 1. Freeman-Carroll plot for thermal degradation of $[\text{CoCPHQ}(\text{H}_2\text{O})_2]_n$ complex.

The kinetic parameter, especially activation energy (E_a) is helpful in assigning the strength of the Co-ordination polymers. The calculated E_a values of Co-ordination polymers indicate that volatile gas is produced in the range of 6.6-9.5 kcalmol⁻¹. The relatively high value of E_a (Table 2) indicates that the ligand is strongly bonded to the metal ion.⁵⁴⁻⁵⁵ Based on the activation energy values, the thermal stability of the Co-ordination polymers in the decreasing order is: Mn > Co > Ni > Zn > Cu.⁵⁶

Antimicrobial Activity of the Bis-ligand and its Co-ordination Polymers

All the novel synthesized compounds and standard drugs were prepared in freshly distilled DMF. The zone of inhibition evaluated was negligible. These data reveal that the Bis-ligand CPHQ and its Co-ordination polymers show weak to good antimicrobial activity (Table 3).

These data are compared with the standard drug streptomycin, the Bis-ligand and its Cu²⁺ and Co²⁺ complexes showed weakly active against the both organisms with zones of inhibition 9-11 mm and 12-14 mm respectively. On the other hand, the Ni²⁺ and Mn²⁺ complexes can be classified as moderately active compared to its ligand with zone of inhibition 15-17 mm while the Zn²⁺ complex exhibited good activity with zone of inhibition 18-20 mm.

Table 3. Antibacterial activities of Bis-ligand (CPHQ) and their Co-ordination polymers.

Ligand/ Co-ordination polymers	Zone of Inhibition (mm)			
	Gram +Ve		Gram -Ve	
	<i>Bacillus subtilis</i>	<i>Staphylococcus aureus</i>	<i>Salmonella typhi</i>	<i>Escherichia coli</i>
CPHQ	11	09	10	10
$[\text{CuCPHQ}(\text{H}_2\text{O})_2]_n$	12	13	14	13
$[\text{CoCPHQ}(\text{H}_2\text{O})_2]_n$	13	12	13	14
$[\text{NiCPHQ}(\text{H}_2\text{O})_2]_n$	16	15	17	16
$[\text{MnCPHQ}(\text{H}_2\text{O})_2]_n$	17	16	16	15
$[\text{ZnCPHQ}(\text{H}_2\text{O})_2]_n$	19	20	18	20
Streptomycin	23	24	24	25
DMF (Control)	00	00	00	00
Bore size	08	08	08	08

The antifungal activity results revealed that the ligand CPHQ and its Co-ordination polymers show weak to good activity (Table 4). Bis-ligand and its Cu²⁺ and Co²⁺ complexes showed weak active with zone of inhibition 12-14 mm. The Ni²⁺ and Zn²⁺ complexes show moderate activity with zone of inhibition 15-17 mm, while the Mn²⁺ complex exhibited good activity with zone of inhibition 18-20 mm.

Table 4. Antifungal activity of Bis-ligand (CPHQ) and their Co-ordination polymers.

Ligand/ Co-ordination polymers	Zone of Inhibition at 1000 ppm (%)			
	<i>Penicillium Expansum</i>	<i>Nigrospora Sp.</i>	<i>Trichothesium Sp.</i>	<i>Rhizopus Nigricum</i>
CPHQ	08	08	09	10
$[\text{CuCPHQ}(\text{H}_2\text{O})_2]_n$	14	13	13	12
$[\text{CoCPHQ}(\text{H}_2\text{O})_2]_n$	12	12	14	13
$[\text{NiCPHQ}(\text{H}_2\text{O})_2]_n$	16	15	17	16
$[\text{MnCPHQ}(\text{H}_2\text{O})_2]_n$	19	18	20	19
$[\text{ZnCPHQ}(\text{H}_2\text{O})_2]_n$	15	17	16	15
Chlotrimazole	24	26	24	25

CONCLUSION

The novel 5-[4-(2-carboxyphenyl carbonyl amino)-phenyl-4-carbonyl methyl amino]-8-hydroxyquinoline (CPHQ) and its octahedral metal (II) complexes (1: 1 metal to ligand ratio) were synthesized and characterized. Further, the Co-ordination polymers were thermally more stable than the ligand. All the Co-ordination polymers showed good antibacterial and antifungal activities compared to CPHQ ligand due to the insertion of the metal ions. The information regarding geometry of the complexes was obtained from their electronic and magnetic moment values. The magnetic moment values indicate an octahedral geometry.

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