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Antimicrobial, spectral and thermal aspects of some novel co-ordination polymers-based on 8-hydroxy quinoline

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ABSTRACT

Looking to the pharmacological importance of 8-hydroxyquinolines,in the present study, a novel bi-dentate ligand N - (3 - (bis (2 - ((8 – hydroxyquinolin – 5 – yl) methoxy) ethyl) amino) – 4 - methoxyphenyl) acetamide (BHQMA) was synthesized by the reaction of N - (3 - (bis (2 - hydroxyethyl) amino) – 4 -methoxyphenyl) acetamide with 5-chloromethyl- 8-hydroxyquinoline hydrochloride. Its co-ordination polymers were also prepared with Mn(II), Fe(II), Co(II), Ni(II) and Cu(II) salts. All the above compounds were investigated by physicochemical analyses, thermogravimetric analysis and spectroscopic techniques. In vitro antimicrobial activity of all synthesized compounds and standard drugs have been evaluated against four strains of bacteria which include two Gram +ve bacteria such as Staphylococcus aureus, Bacillus megaterium and two Gram-ve bacteria such as Es cherichia coli, Proteus vulgaris and one fungi Aspergillus niger.

Introduction

Compounds containing quinoline motif are most widely used as antimalarials, [1] antibacterials [2], antifungals [3] and anticancer agents [4]. Additionally, quinoline derivatives find use in the synthesis of fungicides, virucides, biocides, alkaloids, rubber chemicals and flavoring agents [5]. They are also used as polymers, catalysts, corrosion inhibitors, preservatives, and as solvent for resins and terpenes. Furthermore, these compounds find applications in chemistry of transition-metal catalyst for uniform polymerization and luminescence chemistry [6]. Quinoline derivatives also act as antifoaming agent in refineries [7].

8-Hydroxyquinoline and its derivatives find wide application in coordination chemistry [8], pharmaceutical chemistry [9-11], and materials chemistry [12]. The syntheses and properties of 8- hydroxyquinoline derivatives and their complexes have attracted much research [13].

The research on synthesis and applications of coordination polymers have become a popular field since the past 20 years because of the excellent properties, such as molecular recognition, ion exchange, catalysis, microelectronics, non-linear optics, porous materials, etc. [14–20]. Therefore, it is important to get an object with the expected structure and function through a reasonable choice and control of metal ions, organic ligands and other synthesis conditions. Transition metal ions are easy to coordinate with nitrogen and oxygen atoms, and the coordination numbers are mainly focus on 4 or 6, so it is relatively simple for their coordination geometries [21–27].We prepared novel bis-ligand N - (3 - (bis (2 - ((8 – hydroxyquinolin – 5 – yl) methoxy) ethyl) amino) – 4 - methoxyphenyl) acetamide (BHQMA) which reacts with transition metal ions like Mn(II), Co(II), Ni(II), Cu(II) and Fe(II). The bis-ligand and coordination polymers have been characterized on the basis of physicochemical analyses, thermogravimetric analysis and spectroscopic techniques. The thermal analysis techniques were extensively used in studying the thermal behavior of coordination polymers [28-32].

Experimental

Materials and methods

All the chemicals used, including N-(3- (bis (2-hydroxyethyl) amino)-4-methoxyphenyl) acetamide and 8-hydroxyquinoline, were of analytical grade and were purchased from local markets. They were purified by standard methods prior to use [33]. Nutrientagar and potato dextrose agar were purchased from Hi-media Chemicals, India. Metal(II)-salts (chloride/nitrate/sulfate) of Mn(II), Fe(II), Co(II), Ni(II) and Cu(II) were used in their hydrated form.

Silica gel F_{254} thin-layer chromatographic plates of size 20 × 20 cm were purchased from the E. Merck (India) Limited, Mumbai and used for purity evaluation. The physicochemical analyses (C, H, N) were performed with a model Elemental Vario EL C, H, N elemental analyzer. The infrared spectra (FT-IR) were obtained from KBr pellets in the range 4000–400 cm^{-1} with a Perkin Elmer spectrum GX spectrophotometer (FT–IR) instrument. The ^{1}H NMR spectra was recorded on a Bruker (400 MHz) instrument using DMSO *d*6 as solvent as well as an internal reference standard. Diffuse electronic spectra were recorded on a Beckman DK-2A spectrophotometer using MgO as a reference. Magnetic moments were determined by the Gouy method with mercury tetrathiocyaneto-cobaltate(II), $[HgCo(NCS)_4]$ as calibrant ($\chi_g = 1644 \times 10^{-6}$ cgs units at 20 °C), by Citizen Balance (at room temperature). The diamagnetic correction was made using Pascal's constant [34].The thermogravimetric analyses studies were carried out with a model Perkin Elmer thermogravimetry analyzer at a heating rate of 10 °Cmin^{-1} in air. The metal contents of the complexes were analyzed by EDTA titration after decomposing the organic matter with $HClO_4$, H_2SO_4 and HNO_3 (1: 1.5: 2.5) mixture [35]. The melting point of BHQMA was checked by standard open capillary method and is uncorrected.

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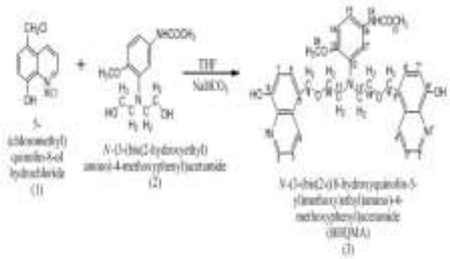
The outline of synthesis of BHQMA and its co-ordination polymers are shown in Scheme 1 and Scheme 2. The physicochemical parameters of co-ordination polymers are summarized in Table 1.

Preparation of bis-ligand (BHQMA)

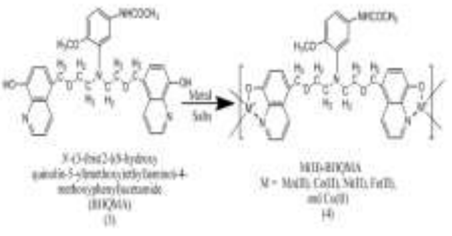
BHQMA was prepared according to the method reported for CMQ-alcohol reaction [36]. To a suspension of 5-chloromethyl-8-hydroxyquinoline hydrochloride (4.6 g, 0.02 mol), N - (3 - (bis (2 - hydroxyethyl) amino) – 4 -methoxyphenyl) acetamide (2.68g, 0.01 mol) in an acetone-water mixture was added. The resulting mixture was refluxed for 3 hr with occasional shaking. The resulting suspension, which contained of a green precipitate was made alkaline with dilute aqueous ammonia and then filtered. The solid product was collected and dried to give BHQMA (72% yield). The formation of the product was confirmed by TLC using $CHCl_3$: CH_3OH (80 : 20) mixture as the mobile phase (R_f 0.53).The product melted with decomposition at above 250 ºC (uncorrected).



Scheme 1. Synthetic route for bis-ligand

Preparation of coordination polymers

A warm solution of metal(II) salt (0.01mol, 2.45g) in 50% aqueous formic acid was added drop by drop with continuous stirring to previously warmed solution of BHQMA (0.005mol, 2.91g) in 20% aqueous formic acid solution. With the proper adjustment of the pH (~8.5) with 50% NH_4OH, the resultant mixture was further digested in a water bath for 4–5 hr and centrifuged. The suspended solid was allowed to settle and collected by filtration, washed with sufficient quantity of distilled water and then with a little hot ethanol and acetonitrile, then dried in vacuum desiccators over anhydrous calcium chloride. The coordination polymers are insoluble in all common organic solvents like methanol, ethanol, chloroform, acetone, benzene and dimethyl sulfoxide. The yield was 80–90%.



Scheme 2. Synthetic route for co-ordination polymers

Antimicrobial activities

The antimicrobial activity was assayed by Cup plate agar diffusion method [37] by measuring inhibition zones in mm. In vitro antimicrobial activity of all synthesized compounds and standard drugs have been evaluated against four strains of bacteria which include two Gram +ve bacteria such as Staphylococcus aureus, Bacillus megaterium and two Gram-ve bacteria such as Es cherichia coli, Proteus vulgaris and one fungi Aspergillus niger.

The antibacterial activity was compared with standard drugs viz. Amoxycillin, Ampicillin, Ciprofloxacin, Erythromycin and antifungal activity was compared with standard drug viz. Griseofulvin.

Antibacterial activity

The purified products were screened for their antibacterial activity by using cup-plate agar diffusion method. The nutrient agar broth prepared by the usual method, was inoculated aseptically with 0.5 mL of 24 hr old subculture of S. aureus, B. megaterium, P. vulgaris, and E. coli in separate conical flasks at 40-50 ^oC and mixed well by gentle shaking. About 25 mL of the contents of the flask were poured and evenly spread in petridish (90 mm in diameter) and allowed to set for two hr. The cups (10 mm in diameter) were formed by the help of borer in agar medium and filled with 0.04 mL (40 pg/mL) solution of sample in DMF.

The plates were incubated at 37 ^oC for 24 hr and the control was also maintained with 0.04 mL of DMF in similar manner and the zones of inhibition of the bacterial growth were measured in millimeter and recorded in Table 2.

Antifungal activity

A. niger was employed for testing antifungal activity by cup-plate agar diffusion method. The culture was maintained on Sub rouse dextrose agar slants. Sterilized Sub rouse dextrose agar medium was inoculated with 72 hr old 0.5 mL suspension of fungal spores in a separate flask. About 25 mL of the inoculated medium was evenly spread in a sterilized petridish and allowed to set for 2 hr. The cups (10 mm in diameter) were punched in petridish and loaded with 0.04 mL (40 pg/mL) of solution of sample in DMF.

The plates were incubated at 30^oC for 48 hr. After the completion of incubation period, the zones of inhibition of growth in the form of diameter in mm were measured. Along the test solution in each petridish one cup was filled up with solvent which acts as control. The zones of inhibition are recorded in Table 2.

Result and discussion

5-Chloromethyl-8-hydroxyquinoline hydrochloride was prepared by chloromethylation of 8-hydroxyquinoline. Considerable difficulties were faced to obtain high purity of CMHQ even after washing the crude CMHQ with concentrated hydrochloric acid and acetone. This might be due to incomplete removal of 8-hydroxyquinoline. Another difficulty was the possibility of substitution reaction during the crystallization with protic solvent. The use of inorganic base catalyst was avoided, such as sodium/potassium bicarbonate, sodium/potassium hydrogen carbonate and sodium hydroxide, as it either leads to a slow reaction or may give 5-hydroxymethyl-8-hydroxyquinoline in quantitative yield [38]. To overcome these difficulties, triethylamine (TEA) was used as scavenger while reacting CMHQ with N - (3 - (bis (2 - hydroxyethyl) amino) – 4 - methoxyphenyl) acetamide to afford intermediate in good yield, which upon acid hydrolysis gave BHQMA in moderate yield.

The synthesized novel N - (3 - (bis (2 - ((8 – hydroxyquinolin – 5 – yl) methoxy) ethyl) amino) – 4 - methoxyphenyl) acetamide (BHQMA) appears as dark green crystals. It has partial solubility in acetone, methanol, ethanol and acetonitrile, while being soluble in polar aprotic solvents like dimethylformamide (DMF), dimethylsulfoxide (DMSO), organic acids and pyridine. All the co-ordination polymers had characteristic color, are stable in air and practically insoluble in water, ethanol, methanol, chloroform and hexane, while low solubility was observed in DMF as well as in DMSO.

The formation of the BHQMA was apparent from the dark blue and yellow spots by visualizing in long- and short-wavelength UV light respectively on the TLC.The results of elemental analyses (C, H, N) of ligand BHQMA and its co-ordination polymers are given in Table 1 and were in good agreement with their predicted molecular formula, showing that the co-ordination polymers have 1 : 2 metal–ligand ratio.

IR Spectra

The spectrum of ligand (BHQMA)(Fig.1) shows a broad band at 3332 cm^{-1} due to –OH stretching vibrations. Weak bands at 2930 cm^{-1} and 2851 cm^{-1} are attributed to asymmetric and symmetric stretching vibrations of methylene groups. The C=N stretching in the ligand is possibly distributed at the absorptions 1706 and 1610 cm^{-1}. On complexation both these peaks show red shift, indicating bonding of nitrogen to the metal. The C-O stretching in the ligand may be traced to the absorptions at 1362 and 1250 cm^{-1}. In the complexes this peaks are shifted to 1345 and 1242 cm^{-1} respectively. This indicating the formation of C-O-M bond. The peak at 1229 cm^{-1} in the ligand is assigned to OH-bending of the phenolic moiety, this is absent in the coordination polymers shown in (Fig.2). The bands at 1485 and 1590 cm^{-1} are due to breathing vibrations in aromatic ring of ligand [39]. In addition to these bands, the spectrum of BHQMA has many characteristic absorption bands which are identical to those that occur in 5,5'-methylene(8- hydroxyquinoline) (MBQ) and bis-ligand [40].

The IR spectrum of BHQMA shows a peak at 1600 cm^{-1} assigned to υ(C=N). On complexation this peak shifted to lower frequencies at 1595 cm^{-1} due to coordination polymer formation [41,42]. The new bands observed in the region 472-455 cm^{-1} and 425 cm^{-1} are probably due to the formation of M-O and M-N bonds respectively [43]. It is thus believed that the oxygen and nitrogen atom of 8-hydroxyquinoline group are coordinated to the metal [44,45]. From the above data the following structure has been proposed for the coordination polymers shown in Scheme 2.

Figure 1. IR spectrum of BHQMA

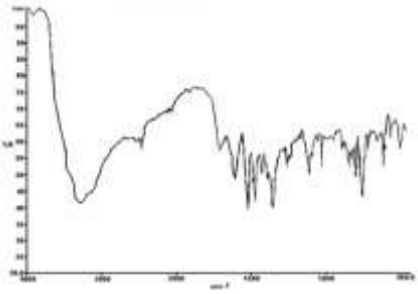


Figure 2. IR spectrum of Cu+2 coordination polymer

^{1}H NMR Analysis

The structure of the ligand (BHQMA) was characterized by ^{1}H NMR spectrum in DMSO-d6 system. The ligand shows a peak at δ = 4.78 ppm (s, 4H, H_9 and $H_{9'}$ protons) is assigned to two –CH_2 protons. Two triplet, one at δ = 3.72 ppm (t, 4H, H_{10} and $H_{10'}$ protons) and secound at 4.16 ppm (t, 4H, H_{11} and $H_{11'}$ protons) is assigned to two ethyl (-CH_2-CH_2-) groups. The peak at δ = 2.04 ppm (s, 3H, -CH_3 protons) is assigned to methyl protons and peak at δ = 11.5 ppm (s, 1H, NH proton) is assigned to aromatic amide proton. The signal at δ = 3.81 ppm (s, 3H, -CH_3 protons) is assigned to aromatic methoxy protons. A sharp peak at δ = 9.5 ppm (s, 2H, -OH protons) is assigned to two aromatic hydroxyl protons [46], which are confirmed by D_2O exchange experiment. Aromatic protons observed at 8.85 (d, 2H, H_2, $H_{2'}$), 7.62 (dd, 2H, H_3 ,$H_{3'}$), 8.50 (d, 2H, H_4, $H_{4'}$), 7.38 (d, 2H, H_6, $H_{6'}$), 7.08 (d, 2H, H_7, $H_{7'}$),6.95(s, 1H, H_{17}),6.76(d, 1H, H_{14}), 6.87(d, 1H, H_{15}).

Diffuse electronic spectral and magnetic properties data

The diffuse electronic spectra of Cu^{+2} complex exhibited two bands at 26237 cm^{-1} due to charge transfer and a broad band having maxima at 15579 cm^{-1} due to the $^2E_g \rightarrow {}^2T_{2g}$ transition. The broadening of the signal might be due to Jahn–Teller distortion. The absorption bands of the diffuse electronic spectra and the value of their magnetic moment favor a tetragonally distorted octahedral geometry around Cu(II) ion [47,48]. Ni^{+2} complex showed three weak absorption bands at 9992, 16110 and 24472 cm^{-1} corresponding to the characteristic transitions $^3A_{2g} \rightarrow {}^3T_{2g}$, $^3A_{2g} \rightarrow {}^3T_{1g}(F)$, $^3A_{2g} \rightarrow {}^3T_{1g}(P)$. Co^{+2} complex exhibited three absorption bands at 9834, 15486 and 22118 cm^{-1}, respectively, due to $^4T_{1g}(F)\rightarrow {}^4T_{2g}(F)$, $^4T_{1g}(F)\rightarrow {}^4A_{2g}(F)$ and $^4T_{1g}(F)\rightarrow {}^4T_{1g}(P)$ transitions. The absorption bands of the diffuse electronic spectra and values of their magnetic moments show an octahedral geometry around Ni(II) and Co(II) ions [49,50]. The spectra of Mn^{+2} complex showed weak bands at 16798, 18435 and 23816 cm^{-1} assigned to the $^6A_{1g} \rightarrow {}^4T_{1g}$,$^6A_{1g} \rightarrow {}^4T_{2g}$,$^6A_{1g} \rightarrow {}^4A_{1g}$,$^4E_g$ transitions and their magnetic moment values suggest an octahedral geometry for the Mn(II) ion. The spectrum for Fe^{+2} shows bands at 18987 and 36735 cm^{-1} assigned to the $^5T_{2g} \rightarrow {}^3E_g$ and $^5T_{2g} \rightarrow {}^3T_{1g}$ transitions, respectively, suggesting its octahedral structure in support of the magnetic moment value around the Fe(II) ion. The results of the magnetic moment value (Table 1) were shown to have octahedral geometry for all the M(II) complexes. Hence, the observed values of magnetic moments and the electronic spectra of M(II) complexes supported octahedral geometry for all the structures [51].

Thermogravimetric analysis

Thermogravimetric analysis (TGA) for the BHQMA and its co-ordination polymers was carried out within the temperature range of 40–900 °C in air at the heating rate of 10 °Cmin^{-1} to establish their compositional differences and to ascertain the nature of associated water molecules. The thermogravimetric curves of all the compounds are presented in Fig. 3. The determined temperature ranges and corresponding percentage mass losses accompanying the changes on heating revealed the following findings.

Securitization of this data envisages that BHQMA follows single-step thermal decomposition. The initial weight loss of 1% might be due to loosely held solvent in BHQMA. The weight loss commences in the range of 150–550 °C up to 74% and further slower degradation takes place up to 900 °C.

Similarly, securitizations of the TGA data of co-ordination polymers of corresponding Mn(II), Fe(II), Co(II), Ni(II) and Cu(II) have shown two-step decomposition. The initial weight loss occurring might be due to the solvent molecules or loosely held moisture trapped inside the co-ordination polymers, whereas the weight loss observed.

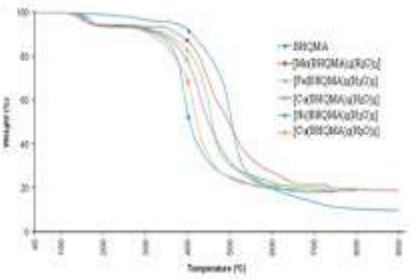


Figure 3.Thermogravimetric curve of BHQMA and its co-ordination polymers.

Antimicrobial activities

Antimicrobial activity of the synthesized compounds and standard drugs is given Table 2. From the Table it is clear that the zones of inhibition area are much larger for the metal chelates than the ligand. The increase in antimicrobial activity is due to faster diffusion of metal complexes as a whole through the cell membrane or due to the combined activity of the metal and ligand [52].

The antimicrobial activity of tested compounds against different strains of bacteria and fungi is shown in Table 2. From Table 2 it can be concluded that all the compounds have displayed maximum activity against P. vulgaris. The compound 4b is highly active against E. coli. The compounds 4b and 4e also showed very good activity against B. megaterium, while compounds 4a and 4c showed good activity against S. aureus. From the data of anti fungal activity it is observed that almost all the compounds are highly active against A. niger except compound 3, which exhibits moderate activity.

As compared to standard drug Ciprofloxacin the compounds are less active, while other drugs have parallel activity.

The substitution of phenyl ring by -OCH_3 have much more effect on the bactericidal and fungicidal activity of complex.

It has been observed that Cu (II) complex have much toxicity. This is expected because the copper salts are mostly used as fungicides.

Such increased activity of the metal complexes can be explained on the basis of Overtone's concept [53] and Tweedy's chelation theory [54].

Most of the compounds inhibit the growth of the above organism, which cause decease in many plants. Hence such type of compounds may find as agricultural and garden bactericides and fungicides.

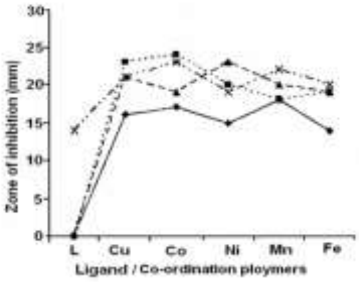


Figure 4. Comparative analysis for Antimicrobial activity of the bis-ligand and coordination polymers

Conclusion

The present paper describes a novel bis-ligand and 8-hydroxyquinoline moieties. The bis-ligand (BHQMA) affords coordination polymers with metal ions. The coordination polymers are thermally stable then ligand. All the coordination polymers have good antimicrobial activity relative to the ligand due to the insertion of metal ions.

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Table 1. Physicochemical parameter of the BHQMA ligand and its co-ordination polymers

Sr	Empirical Formula	Mol. Wt.	Color	Yield (%)	m.p. (°C)	Elemental analysis, % found				μ_{eff} B.M.
						C (Calcd)	H (Calcd)	N (Calcd)	M (Calcd)	
3	$C_{33}H_{34}O_6N_4$	582	Dark Green	72	>250	68.01 (68.04)	5.83 (5.84)	9.65 (9.62)	-	-
4a	$C_{33}H_{32}O_6N_4Cu_2.2H_2O$	744	Dark Green	81	>300	53.24 (53.22)	4.31 (4.30)	7.54 (7,52)	17.19 (17.20)	2.14
4b	$C_{33}H_{32}O_6N_4Ni_2.2H_2O$	736	Dark Red	87	>300	53.77 (53.80)	4.36 (4.34)	7.61 (7.60)	16.32 (16.30)	3.32
4c	$C_{33}H_{32}O_6N_4Co_2.2H_2O$	734	Dark Green	84	>300	53.98 (53.95)	4.34 (4.35)	7.60 (7.62)	16.10 (16.07)	4.64
4d	$C_{33}H_{32}O_6N_4Fe_2.2H_2O$	728	Dark Brown	90	>250	54.38 (54.39)	4.40 (4.39)	7.70 (7.69)	15.39 (15.38)	5.42
4e	$C_{33}H_{32}O_6N_4Mn_2.2H_2O$	726	Brown	83	>300	54.56 (54.54)	4.42 (4.40)	7.72 (7.71)	15.13 (15.15)	5.63

Table 2. Microbiological evaluation of compounds

Organism	Compounds						Standard drugs				
	3	4a	4b	4c	4d	4e	Ampicillin	Amoxycillin	Ciprofloxacin	Erythromycin	Griseofulvin
E. coil	19	22	25	19	18	24	18	16	25	24	0
P. vulgaris	21	20	28	20	21	21	23	21	29	17	0
B. mega	16	18	20	24	22	26	21	24	21	12	0
S. aureus	16	24	17	23	17	19	27	28	23	24	0
A. niger	20	21	23	19	23	22	0	0	0	0	22