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**DRUG SYNTHESIS**

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**SYNTHESIS, CHARACTERIZATION AND ANTIBACTERIAL STUDIES  
ON MIXED LIGAND COPPER COMPLEXES WITH POLYDENTATE LIGANDS**

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**Abstract:** Mixed ligand Cu(II) complexes of the type  $[M(Q)(L)] \cdot 2H_2O$  have been synthesized using 8-hydroxyquinoline (HQ) as a primary ligand and N- and/or O-donor amino acids (HL) such as L-threonine, L-proline, L-hydroxyproline, L-isoleucine and L-serine as secondary ligands. The metal complexes have been characterized on the basis of elemental analysis, electrical conductance, room temperature magnetic susceptibility measurements, spectral and thermal studies. The electrical conductance studies of the complexes in DMSO (dimethyl sulfoxide) in  $10^{-3}$  M concentration indicate their non-electrolytic nature. Room temperature magnetic susceptibility measurements revealed paramagnetic nature of the complexes. Electronic absorption spectra of the complexes show intra-ligand, charge transfer transitions and d-d transitions. The thermal analysis data of the complexes indicate the presence of crystallized water molecules. The agar cup method and tube dilution method have been used to study the antibacterial activity of the complexes against the pathogenic bacteria *S. aureus*, *C. diphtheriae*, *P. aeruginosa* and *E. coli*. The results have been compared with those of tetracycline, which was screened simultaneously and indicated mild antibacterial activity of the complexes.

**Key words:** Mixed ligand copper complexes; synthesis, antibacterial study

Many researchers have studied characterization, antimicrobial and toxicological activity of mixed ligand complexes of transition metals (1–6). The role of mixed ligand complexes in biological process has been well recognized (7, 8). It has been found that a majority of the metal complexes with 8-hydroxyquinoline possess biological activity (9–11). Amino acids are well known for their tendency to form complexes with metals having biological significance and metabolic enzymatic activities (12). Anti-tumor activity of some mixed ligand complexes has also been reported (13, 14). The antibacterial and anti-fungal properties of a range of copper(II) complexes have been evaluated against several pathogenic bacteria and fungi (15–18).

Therefore, it was considered to study the complexation and to determine the biological activity of copper complexes. The present paper reports synthesis, characterization and antibacterial studies of mixed ligand Cu(II) complexes prepared with 8-hydroxyquinoline (HQ) as a primary ligand and amino acids (HL) such as L-threonine, L-proline, L-hydroxyproline, L-isoleucine and L-serine as sec-

ondary ligands. The metal complexes have been characterized by elemental analysis and various physico-chemical techniques such as molar conductance, magnetic susceptibility, electronic spectra, IR spectra and thermal studies.

**EXPERIMENTAL****Materials**

Analytical grade copper(II) chloride dihydrate was used as such without further purification. L-threonine, L-proline, L-hydroxyproline, L-isoleucine, L-serine and 8-hydroxyquinoline were obtained from S.D. Fine Chemicals, Mumbai, India. Solvents like, ethanol, dimethyl sulfoxide and laboratory grade chemicals, whenever used, were distilled and purified according to standard procedures (19, 20).

**Preparation of mixed ligand complexes**

Mixed ligand Cu(II) complexes were prepared from copper(II) chloride dihydrate, 8-hydroxyquinoline (HQ) as a primary ligand and different

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amino acids (HL) such as L-threonine, L-proline, L-hydroxyproline, L-isoleucine and L-serine as secondary ligands.

To an aqueous solution (10 cm<sup>3</sup>) of copper(II) chloride dihydrate (170 mg, 1 mmol), ethanolic solution (10 cm<sup>3</sup>) of 8-hydroxyquinoline (145 mg, 1 mmol) was added. The mixture was stirred and kept in a boiling water bath for 10 min. To this hot solution, an aqueous solution (10 cm<sup>3</sup>) of amino acids (1 mmol) was added with constant stirring. The mixture was again heated in a water bath. The complexes were obtained by raising pH of the reaction mixture by adding diluted ammonia solution. The mixture was cooled and solid complex obtained was filtered, washed with water followed by ethanol. The complexes thus prepared were dried under vacuum.

### Instrumentation

The complexes were analyzed for C, H, and N contents on Thermo Finnigan Elemental Analyzer, Model No. FLASH EA 1112 Series at Department of Chemistry, I.I.T., Mumbai. Metal content was estimated complexometrically by standard procedure (21, 22).

The molar conductance values were measured in DMSO (10<sup>-3</sup> M) on an Equip-tronics Autoranging Conductivity Meter Model No. EQ-667 with a dip type conductivity cell fitted with platinum electrodes (cell constant = 1.0 cm<sup>-1</sup>).

The room temperature magnetic susceptibility measurements of the complexes reported in the present study were made by the Guoy's method using Hg[Co(SCN)<sub>4</sub>] as calibrant at Department of Chemistry, I.I.T., Mumbai.

The electronic absorption spectra of all the complexes in DMSO solution (10<sup>-3</sup> M) in the ultraviolet and visible region were recorded on Shimadzu UV/VIS-160 spectrophotometer at GNIRD, Mumbai.

Infrared spectra of all the ligands and their metal complexes were recorded in KBr discs on a Perkin-Elmer FT-IR spectrophotometer Model 1600 in the region 4000–400 cm<sup>-1</sup> at Department of Chemistry, I.I.T., Mumbai.

The thermogravimetric (TG) and differential thermal analysis (DTA) measurements were carried out in controlled nitrogen atmosphere on a Perkin-Elmer Diamond TG-DTA Instrument at the Department of Chemistry, I.I.T., Mumbai by recording the change in weight of the complexes on increasing temperature up to 900°C at heating rate of 10°C per min.

### Antibacterial screening

#### Agar cup method

In the agar cup method, a single compound can be tested against number of organisms or a given organism against different concentrations of the same compound. The method was found suitable for semisolid or liquid samples and was used in the present work. In the agar cup method, a plate of sterile nutrient agar with the desired test strain was poured to a height of about 5 mm, allowed to solidify and a single cup of about 8 mm diameter was cut from the center of the plate with a sterile cork borer. Thereafter, the cup was filled with the sample solution of known concentration and the plate was incubated at 37°C for 24 h. The extent of inhibition of growth from the edge of the cup was considered as a measure of the activity of the given compound. By using several plates simultaneously, the activities of several samples were quantitatively studied.

#### Tube dilution method

The test compound (10 mg) was dissolved in DMSO (10 cm<sup>3</sup>) so as to prepare a stock solution of concentration 1000 µg/mL. From this stock solution, aliquots of 50 to 1000 µg/mL were obtained in test broth.

The test compounds were subjected to *in vitro* screening against *Staphylococcus aureus*, *Corynebacterium diphtheriae*, *Pseudomonas aeruginosa* and *Escherichia coli* using Muller Hinton broth as the culture medium.

Bacterial inoculums were prepared in sterilized Muller Hinton broth and incubated for 4 h at 37°C. They were dispersed (5 cm<sup>3</sup>) in each borosilicate test tube (150 × 20 mm). The test sample solution was added in order to attain a final concentration as 50 to 1000 µg/mL. The bacterial inoculums 0.1 cm<sup>3</sup> of the desired bacterial strain (*S. aureus*, *C. diphtheriae*, *P. aeruginosa* and *E. coli*) containing 10<sup>6</sup> bacteria/cm<sup>3</sup> were inoculated in the tubes. The tubes were incubated at 37°C for 24 h and then examined for the presence or absence of the growth of the test organisms.

The lowest concentration which showed no visible growth was noted as minimum inhibitory concentration (MIC).

## RESULTS AND DISCUSSION

### Characterization of metal complexes

The synthesis of mixed ligand Cu(II) complexes may be represented as follows:

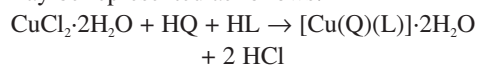


Table 1. Empirical formula, molecular weight, color, decomposition temperature and pH of the copper complexes studied.

No.	Complex	Empirical formula	Molecular weight	Color	Decomposition temperature (°C)	pH
1	[Cu(Q)(Thr)]·2H <sub>2</sub> O	CuC <sub>13</sub> H <sub>18</sub> O <sub>6</sub> N <sub>2</sub>	361.54	Green	254	6.99
2	[Cu(Q)(Pro)]·2H <sub>2</sub> O	CuC <sub>14</sub> H <sub>18</sub> O <sub>5</sub> N <sub>2</sub>	357.54	Yellowish green	258	6.98
3	[Cu(Q)(Hpro)]·2H <sub>2</sub> O	CuC <sub>14</sub> H <sub>18</sub> O <sub>6</sub> N <sub>2</sub>	373.54	Yellowish green	276	7.01
4	[Cu(Q)(Iso)]·2H <sub>2</sub> O	CuC <sub>15</sub> H <sub>22</sub> O <sub>5</sub> N <sub>2</sub>	373.54	Dark green	276	6.99
5	[Cu(Q)(Ser)]·2H <sub>2</sub> O	CuC <sub>12</sub> H <sub>16</sub> O <sub>6</sub> N <sub>2</sub>	347.54	Green	276	7.00

Q represents the deprotonated primary ligand – 8-hydroxyquinoline, whereas Thr, Pro, Hpro, Iso, and Ser represent deprotonated secondary ligands: L-threonine, L-proline, 4-hydroxy-L-proline, L-isoleucine and L-serine, respectively.

Table 2. Elemental analysis data, molar conductance and magnetic moments of copper complexes.

No.	Complex	Elemental analysis Found (Calcd.)				Molar conductance (Mhos cm <sup>2</sup> mol <sup>-1</sup> )	$\mu_{\text{eff}}$ (B.M.)
		% M	% C	% H	% N		
1	[Cu(Q)(Thr)]·2H <sub>2</sub> O	17.55 (17.57)	43.10 (43.14)	4.93 (4.97)	7.73 (7.74)	0.028	1.91
2	[Cu(Q)(Pro)]·2H <sub>2</sub> O	17.75 (17.77)	46.99 (46.98)	5.04 (5.03)	7.81 (7.83)	0.013	1.72
3	[Cu(Q)(Hpro)]·2H <sub>2</sub> O	17.11 (17.01)	44.99 (44.97)	4.79 (4.81)	7.47 (7.49)	0.012	1.91
4	[Cu(Q)(Iso)]·2H <sub>2</sub> O	17.05 (17.01)	48.19 (48.18)	5.85 (5.88)	7.49 (7.49)	0.014	1.88
5	[Cu(Q)(Ser)]·2H <sub>2</sub> O	18.24 (18.28)	55.22 (55.24)	4.62 (4.60)	8.01 (8.05)	0.022	1.99

Abbreviations see Table 1.

where, HQ is 8-hydroxyquinoline and HL is an amino acid.

All the complexes are colored, non-hygroscopic and thermally stable solids (Table 1), indicating a strong metal-ligand bond. The complexes are insoluble in common organic solvents such as ethyl alcohol, acetone, etc., but are fairly soluble in DMSO. The elemental analysis data (Table 2) of metal complexes are consistent with their general formulation as 1:1:1, mixed ligand complexes of the type [Cu(Q)(L)]·2H<sub>2</sub>O. The molar conductance values of the complexes in DMSO at 10<sup>-3</sup> M concentration are very low (< 1) indicating their non-electrolytic nature (23).

#### Magnetic studies

The magnetic moments of the metal complexes were calculated from the measured magnetic susceptibilities after employing diamagnetic corrections and revealed their paramagnetic nature. The observed values for effective magnetic moment

( $m_{\text{eff}}$ ) in BM, reported in Table 2, suggest the square planar geometry for copper complexes. The magnetic moments of the compounds investigated support the conclusions.

#### Electronic absorption spectra

The electronic spectra of the metal complexes in DMSO were recorded in the UV-visible region. The spectra show three transitions in the range 35971–37037 cm<sup>-1</sup>, 29586–29674 cm<sup>-1</sup> and 25253–25641 cm<sup>-1</sup> ascribed to  $\pi \rightarrow \pi^*$ ,  $n \rightarrow \pi^*$  and the charge transfer transitions from the ligands to the metal, respectively.

#### Infra-red spectra

The FTIR spectra of the metal complexes were recorded in KBr discs over the range 4000–400 cm<sup>-1</sup>. These spectra were complex due to the presence of numerous bands with varying intensities, making the interpretation task quite difficult. However, an attempt has been made to assign some of the impor-

Table 3. Thermal data of copper complexes.

No.	Complex	Temperature range for loss of water molecules (°C)	% Weight loss due to water		Temperature range for loss of 8HQ and amino acid (°C)	Weight loss due to 8HQ and amino acid	
			Found	Calcd.		Found	Calcd.
1	[Cu(Q)(Thr)]·2H <sub>2</sub> O	31–90	10.50	09.95	254–545	71.53	72.45
2	[Cu(Q)(Pro)]·2H <sub>2</sub> O	31–85	10.50	10.06	258–540	72.21	71.50
3	[Cu(Q)(Hpro)]·2H <sub>2</sub> O	31–58	10.00	09.61	276–527	73.39	73.35
4	[Cu(Q)(Iso)]·2H <sub>2</sub> O	27–54	10.50	09.63	276–554	72.89	73.35
5	[Cu(Q)(Ser)]·2H <sub>2</sub> O	36–67	10.76	10.35	276–540	72.63	71.25

Abbreviations see Table 1.

Table 4. Antibacterial activity (mm) of copper complexes by agar cup method.

No.	Complex	Test			
		<i>S. aureus</i>	<i>C. diphtheriae</i>	<i>P. aeruginosa</i>	<i>E. coli</i>
1	[Cu(Q)(Thr)]·2H <sub>2</sub> O	26	12	24	12
2	[Cu(Q)(Pro)]·2H <sub>2</sub> O	12	12	16	11
3	[Cu(Q)(Hpro)]·2H <sub>2</sub> O	26	18	22	12
4	[Cu(Q)(Iso)]·2H <sub>2</sub> O	20	12	24	12
5	[Cu(Q)(Ser)]·2H <sub>2</sub> O	28	12	18	11
6.	Tetracycline	30	25	18	18

Abbreviations see Table 1.

Table 5. MIC (mg/mL) data of copper complexes.

No.	Complex	<i>S. aureus</i>	<i>C. diphtheriae</i>	<i>P. aeruginosa</i>	<i>E. coli</i>
1	[Cu(Q)(Thr)]·2H <sub>2</sub> O	400	800	400	800
2	[Cu(Q)(Pro)]·2H <sub>2</sub> O	800	800	600	800
3	[Cu(Q)(HPro)]·2H <sub>2</sub> O	400	600	400	600
4	[Cu(Q)(Iso)]·2H <sub>2</sub> O	600	800	400	800
5	[Cu(Q)(Ser)]·2H <sub>2</sub> O	400	800	600	600
6	Tetracycline	1.5	2.0	8.0	4.0

Abbreviations see Table 1.

tant bands on the basis of reported infrared spectra of several N- and/or O-donor ligands, 8-hydroxyquinoline and their metal complexes (24–27).

An important feature of infrared spectra of the metal complexes is the absence of band at ~3440 cm<sup>-1</sup> due to the O-H stretching vibration of the free O-H group of HQ. This observation leads to the conclusion that complex formation takes place by deprotonation of the hydroxyl group of HQ moiety. A strong ν(CO) band observed in the range 1112–1111 cm<sup>-1</sup> in the spectra of the complexes, indicates the presence of the 8-hydroxyquinolate group in the complexes coordinating through its nitrogen and oxygen atoms as uninegative bidentate

ligand. The ν(C=N) mode observed at 1580 cm<sup>-1</sup> in the spectrum of free HQ ligand is found to be shifted to lower wave number i.e., 1500–1470 cm<sup>-1</sup> in the spectra of the complexes, suggesting coordination through the tertiary nitrogen donor of HQ. The in-plane and out-of-plane deformation modes observed at ~500 cm<sup>-1</sup> and ~780 cm<sup>-1</sup>, respectively, in the spectrum of HQ are shifted to higher wave numbers ~517 cm<sup>-1</sup> and in the range 788–782 cm<sup>-1</sup>, respectively, confirming co-ordination through the nitrogen atom of HQ with the metal ion (9, 10, 24).

A broad band observed in the region 3432–3200 cm<sup>-1</sup> due to asymmetric and symmetric O–H stretching modes and a weak band in the range

1579–1578  $\text{cm}^{-1}$  due to H–O–H bending vibrations indicate the presence of a coordinated water molecule (28–30), further confirmed by thermal studies. The N–H asymmetric and N–H symmetric vibrations observed at  $\sim 3040$  and  $\sim 2960$   $\text{cm}^{-1}$ , respectively, in the free amino acids are shifted to higher wave numbers i.e., in the range 3142–3085  $\text{cm}^{-1}$  and 3085–3054  $\text{cm}^{-1}$ , respectively, in the spectra of the complexes, suggesting co-ordination of the amino group through nitrogen with the metal ion. The  $\nu_{\text{asymmetric}}$  (COO<sup>-</sup>) band of the free amino acids i.e.,  $\sim 1590$   $\text{cm}^{-1}$  is shifted to higher wave number, i.e., in the range 1618–1600  $\text{cm}^{-1}$  and the  $\nu_{\text{symmetric}}$  (COO<sup>-</sup>) mode observed at  $\sim 1400$   $\text{cm}^{-1}$  in the spectra of free amino acids is found to be shifted to lower wave number i.e., 1378–1377  $\text{cm}^{-1}$ , in the spectra of complexes indicating the co-ordination of the carboxylic acid group *via* oxygen with the metal ion. The difference ( $\nu_{\text{asymmetric}} - \nu_{\text{symmetric}}$ ) is in the range 241–223  $\text{cm}^{-1}$  indicating that the M–O bond is purely covalent (31, 32). The C–N symmetrical stretching frequency observed at  $\sim 950$   $\text{cm}^{-1}$  in the spectra of amino acids is found to be shifted to lower wave numbers i.e., 921–905  $\text{cm}^{-1}$  in the spectra of the complexes, confirming coordination through the amino group of the amino acids.

Some new bands of weak intensity observed in the regions around 639–607  $\text{cm}^{-1}$  and 410  $\text{cm}^{-1}$  may be ascribed to the M–O and M–N vibrations, respectively (10, 33, 34). It may be noted that these vibrational bands are absent in the infra-red spectra of HQ as well as amino acids.

### Thermal studies

The TG and DTA studies of the complexes have been recorded in the nitrogen atmosphere at the constant heating rate of 10°C/min.

The TG of the complexes shows that they are thermally quite stable to varying degree. The complexes show gradual loss in weight due to decomposition by fragmentation with increasing temperature as presented in Table 3. All the complexes show similar behavior in TG and DTA studies. The thermograms of these complexes show the loss in weight corresponding to two water molecules in the temperature range 27–90°C, followed by simultaneous weight loss in the range 254–554°C due to amino acid and 8-hydroxyquinoline moieties.

The DTA of the complexes display an endothermic peak in the range 27–90°C, which indicates the presence of crystallized water molecules.

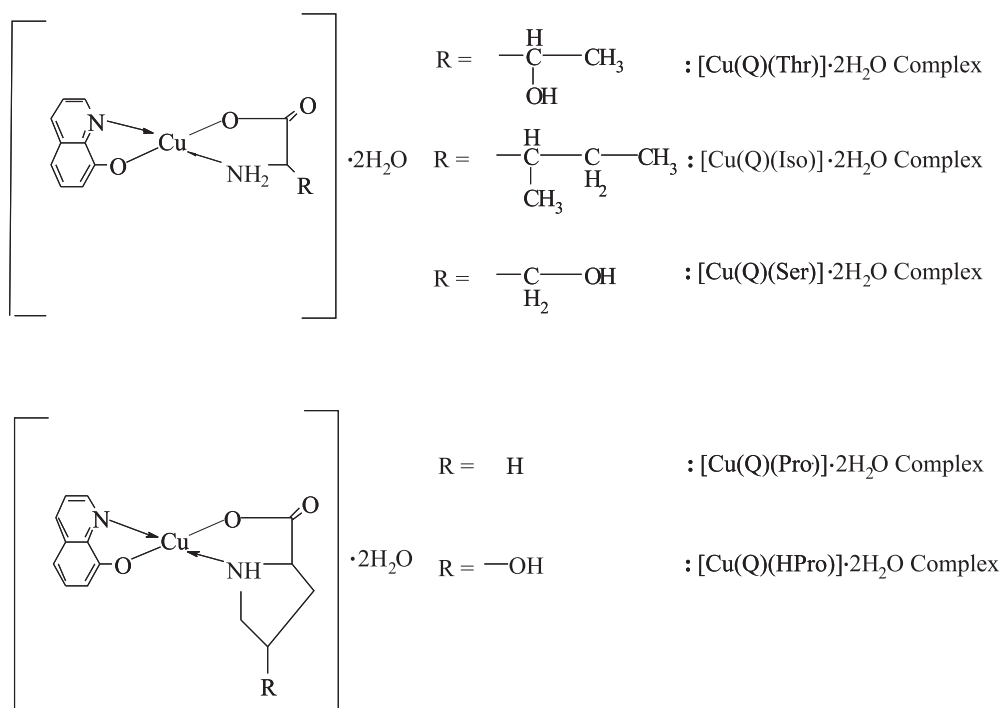


Figure 1. The proposed structures of copper complexes

As the temperature is raised, the DTA curves show a broad exotherm in the range 254–554°C attributed to simultaneous decomposition of amino acid and 8-hydroxyquinoline moieties present in the complexes. The formation of a broad exotherm is possibly due to simultaneous decomposition of ligand moieties and their subsequent oxidation to gaseous products like CO<sub>2</sub> and H<sub>2</sub>O (29).

Like most of the metal organic complexes, these complexes also decomposes to a fine powder of metal oxide i.e., CuO. The constant weight plateau in TG above 600°C indicates completion of the reaction. The CuO formed was confirmed by X-ray diffraction pattern of the decomposed product (29).

### Biological studies

All the metal complexes were screened against *Staphylococcus aureus*, *Corynebacterium diphtheriae*, *Pseudomonas aeruginosa* and *Escherichia coli*.

The studies based on agar cup method revealed that the complexes are sensitive against *S. aureus* and *P. aeruginosa* and less sensitive against *C. diphtheriae* and *E. coli* (Table 4).

The minimum inhibitory concentration (MIC) of complexes (Table 5) ranges between 400–800 µg/mL. The complexes are found to be more active against *S. aureus* and *P. aeruginosa* as compared to *C. diphtheriae* and *E. coli*. As compared to standard antibacterial compound, tetracycline, the complexes show minor activity against selected strains of microorganisms (29, 35–38). Minor activity of these complexes is due to bulky structure of the complexes.

On the basis of the physico-chemical studies, the bonding and structure for the copper complexes may be represented as shown in Figure 1.

### CONCLUSIONS

The higher decomposition temperatures of the complexes indicate a strong metal-ligand bond and electrical conductance studies show non-electrolytic nature of the complexes. Magnetic studies indicate paramagnetic nature of the complexes. Electronic absorption spectra of the complexes show intra-ligand and charge transfer transitions. IR spectra show bonding of the metal ion through N- and O-donor atoms of the two ligands. Thermal analysis confirms the presence of crystallized water molecules. On the basis of above results, square planar structure is proposed for copper complexes under study.

The antibacterial study shows that complexes are found to be more active against *S. aureus* and *P.*

*aeruginosa* as compared to *C. diphtheriae* and *E. coli*. Compared to standard antibacterial compound, tetracycline, the complexes show minor activity against selected strains of microorganisms.

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