



Faculty of Education
Ain Shams University

**Synthesis and spectral studies of transition metal
complexes of some heterocyclic hydrazones derived
from 4,6-diacetylresorcinol**

A Thesis Submitted by

Mona Ahmed Ahmed Mohamed Kishk

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Under Supervision

Prof. Dr. Saied Mohamed Khalil

Dr. Magdy Shebl Saleh El-Sayed

Dr. Mosad Abd-Elrahman Elghamry

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Board of Advisors

Approved by

1) Prof. Dr. Saied M. Khalil

Professor of Inorganic Chemistry,
Faculty of Education, Ain Shams University.

2) Ass. Prof. Dr. Magdy Shebl Saleh El-Sayed

Associate Professor of Inorganic Chemistry,
Faculty of Education, Ain Shams University.

3) Dr. Mossad Abd El-Rahman Elgamry

Lecturer of Inorganic Chemistry,
Faculty of Education, Ain Shams University.

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Mono- and binuclear copper(II) complexes of new hydrazone ligands derived from 4,6-diacetylresorcinol: Synthesis, spectral studies and antimicrobial activity



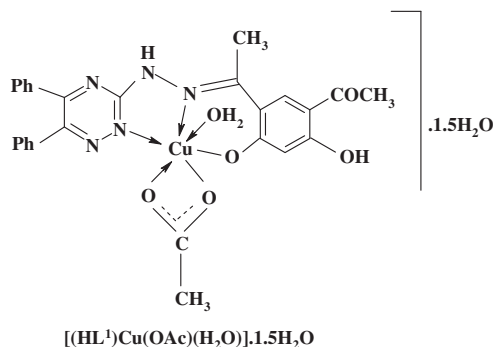
Magdy Shebl*, Mosad A. El-ghamry, Saied M.E. Khalil, Mona A.A. Kishk

Department of Chemistry, Faculty of Education, Ain Shams University, Roxy, Cairo, Egypt

HIGHLIGHTS

- Two new hydrazone ligands were synthesized and characterized.
- Copper(II) complexes were synthesized and characterized by analytical and spectral methods.
- The spin Hamiltonian parameters of some complexes were calculated and discussed.
- The complexes exhibited octahedral and square planar geometrical arrangements.
- The ligands and some complexes showed antimicrobial activity.

GRAPHICAL ABSTRACT



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ABSTRACT

Two new hydrazone ligands, H₂L¹ and H₂L², were synthesized by the condensation of 4,6-diacetylresorcinol with 3-hydrazino-5,6-diphenyl-1,2,4-triazine and isatin monohydrazone, respectively. The structures of the ligands were elucidated by elemental analyses, IR, ¹H NMR, electronic and mass spectra. Reactions of the ligands with several copper(II) salts, including AcO⁻, NO₃⁻, SO₄²⁻, Cl⁻ and Br⁻ afforded mono- and binuclear metal complexes. Also, the ligands were allowed to react with Cu(II) ion in the presence of a secondary ligand (L') [N,O-donor; 8-hydroxyquinoline, N,N-donor; 1,10-phenanthroline or O,O-donor; benzoylacetone]. Characterization and structure elucidation of the prepared complexes were achieved by elemental and thermal analyses, IR, electronic, mass and ESR spectra as well as conductivity and magnetic susceptibility measurements. The ESR spin Hamiltonian parameters of some complexes were calculated. The spectroscopic data showed that the H₂L¹ ligand acts as a neutral or monobasic tridentate ligand while the H₂L² ligand acts as a bis(monobasic tridentate) ligand. The coordination sites with the copper(II) ion are phenolic oxygen, azomethine nitrogen and triazinic nitrogen (H₂L¹ ligand) or isatinic oxygen (H₂L² ligand). The metal complexes exhibited octahedral and square planar geometrical arrangements depending on the nature of the anion. The ligands and some metal complexes showed antimicrobial activity.

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Introduction

Hydrazones and their metal complexes form an interesting class of compounds which find extensive applications in antibacterial,

* Corresponding author. Tel.: +20 233120546.

E-mail address: magdy_shebl@hotmail.com (M. Shebl).

antifungal and antitumor agents [1–3]. Also, they are used in extraction of some metal ions using different buffer solutions [4], micro determination of metal ions [4], determination of titanium in bauxite, Portland cement, amphibolites and granites [5].

Triazine is an important class of heterocyclic compounds found in many synthetic and natural products with a wide range of

biological activities, such as adenosine receptor antagonist [6], antiameobic [7], antimalarial [8], antiviral [9], antitubercular [10] and carbonic anhydrase inhibitor [11].

Isatin is a versatile lead molecule for designing potential bioactive agents and its derivatives were reported to possess a broad spectrum of antiviral activities [12]. Ligands containing isatin moiety are known to possess a wide range of pharmacological properties that include antibacterial, antifungal [13], anticonvulsant [14] and anti-HIV [15] activities.

The bifunctional carbonyl compound; 4,6-diacetylresorcinol (DAR) serves as precursor for the construction of different polydentate ligands [16–20]. In our previous studies, metal complexes of polydentate ligands derived from 4,6-diacetylresorcinol [21–27] have been synthesized and fully characterized. The most motivating features of these ligands are the possibility of use them to synthesize polynuclear complexes with different modes of bonding. Also, mixed-ligand complexes including 4,6-diacetylresorcinol as a primary ligand have been studied [28–30].

On the basis of stated facts, the present work is an extension to our work and is devoted to the synthesis of new hydrazone ligands, H_2L^1 and H_2L^2 , by the condensation of 4,6-diacetylresorcinol with 3-hydrazino-5,6-diphenyl-1,2,4-triazine and isatin monohydrazone, respectively in the molar ratio 1:2 (DAR:hydrazino). The ligational behavior of the new ligands towards different copper(II) salts was investigated. The structures of the ligands and their metal complexes were characterized by elemental and thermal analyses, IR, 1H NMR, electronic, ESR and mass spectra as well as conductivity and magnetic susceptibility measurements at room temperature. The biological activity of the ligands and their complexes was screened against selected kinds of bacteria and fungi.

Experimental

Materials

4,6-Diacetylresorcinol was prepared as cited in the literature [16]. 3-Hydrazino-5,6-diphenyl-1,2,4-triazine was prepared according to Ref. [31] and isatin monohydrazone was prepared according to Ref. [32]. Benzil, glacial acetic acid, thiosemicarbazide, hydrazine hydrate and isatin were BDH or Merck products. Metal salts, lithium hydroxide, 8-hydroxyquinoline (8-HQ), 1,10-phenanthroline (Phen), benzoylacetone (Bac), EDTA disodium salt, ammonium hydroxide, murexide and nitric acid were either Aldrich,

Table 1
Analytical and physical data of the hydrazone, H_2L^1 and H_2L^2 , ligands and their copper(II) complexes.

No.	Reaction	Complex M.F. [F. Wt]	Color	Yield ^a (%)	M.P. (°C)	Elemental analysis, % found/(calc.)				
						C	H	N	Cl/S	M
	H_2L^1	$C_{25}H_{21}N_5O_3$ [439.48]	Pale brown	89	>300	68.46 (68.33)	4.7 (4.82)	16.1 (15.94)	–	–
(1)	$H_2L^1 + Cu(OAc)_2 \cdot H_2O$	$[(HL^1)Cu(OAc)(H_2O)] \cdot 1.5H_2O$ $C_{27}H_{28}N_5O_{7.5}Cu$ [606.1]	Brown	83	>300	53.57 (53.51)	4.97 (4.66)	11.8 (11.55)	–	10.4 (10.48)
(2)	$H_2L^1 + Cu(NO_3)_2 \cdot 2.5H_2O$	$[(HL^1)Cu(H_2O)]NO_3$ $C_{25}H_{22}N_6O_7Cu$ [582.04]	Dark green	84	>300	51.66 (51.59)	3.8 (3.81)	14.56 (14.44)	–	11.0 (10.92)
(3)	$H_2L^1 + CuSO_4 \cdot 5H_2O$	$[(H_2L^1)Cu(SO_4)(H_2O)_2] \cdot 1.5H_2O$ $C_{25}H_{28}N_5O_{10.5}SCu$ [662.14]	Brown	50	>300	45.42 (45.35)	4.0 (4.26)	10.47 (10.58)	4.6 (4.84)	9.4 (9.6)
(4)	$H_2L^1 + CuCl_2 \cdot 2H_2O$	$[(HL^1)CuCl] \cdot 1.5H_2O$ $C_{25}H_{23}N_5O_{4.5}ClCu$ [564.49]	Green	75	281	52.9 (53.19)	4.2 (4.11)	12.29 (12.41)	6.0 (6.28)	11.1 (11.26)
(5)	$H_2L^1 + CuBr_2$	$[(HL^1)Cu(H_2O)_3]Br \cdot H_2O$ $C_{25}H_{28}N_5O_7BrCu$ [653.98]	Dark green	60	260	45.84 (45.92)	4.1 (4.32)	10.88 (10.71)	–	9.5 (9.72)
(6)	$H_2L^1 + Cu(OAc)_2 \cdot H_2O + 8-HQ$	$[(HL^1)Cu(8-HQ)(H_2O)]$ $C_{34}H_{28}N_6O_5Cu$ [664.19]	Chocolate brown	96	263	61.47 (61.49)	4.0 (4.25)	12.9 (12.65)	–	9.4 (9.57)
(7)	$H_2L^1 + Cu(OAc)_2 \cdot H_2O + Phen$	$[(HL^1)Cu(OAc)(Phen)]$ $C_{39}H_{31}N_7O_5Cu$ [741.27]	Dark brown	71	267	63.4 (63.19)	3.9 (4.22)	13.5 (13.23)	–	8.4 (8.57)
(8)	$H_2L^1 + Cu(OAc)_2 \cdot H_2O + Bac$	$[(HL^1)Cu(H_2O)(Bac)] \cdot 2H_2O$ $C_{35}H_{35}N_5O_8Cu$ [717.24]	Dark brown	71	>300	58.3 (58.61)	4.7 (4.92)	9.9 (9.76)	–	8.7 (8.86)
	H_2L^2	$C_{26}H_{20}N_6O_4$ [480.49]	Brown	94	>300	64.8 (64.99)	4.5 (4.2)	17.2 (17.49)	–	–
(9)	$H_2L^2 + Cu(OAc)_2 \cdot H_2O$	$[(L^2)Cu_2(OAc)_2(EtOH)_2] \cdot 0.5H_2O \cdot 0.5EtOH$ $C_{35}H_{40}N_6O_{11}Cu_2$ [847.83]	Dark brown	59	>300	49.6 (49.58)	4.5 (4.76)	9.79 (9.91)	–	14.8 (14.99)
(10)	$H_2L^2 + Cu(NO_3)_2 \cdot 2.5H_2O$	$[(L^2)_2Cu_2] \cdot EtOH$ $C_{54}H_{42}N_{12}O_9Cu_2$ [1130.10]	Chocolate brown	17	>300	57.37 (57.39)	3.77 (3.75)	14.6 (14.87)	–	11.20 (11.25)
(11)	$H_2L^2 + CuSO_4 \cdot 5H_2O$	$[(L^2)_2Cu_2] \cdot 2H_2O$ $C_{52}H_{40}N_{12}O_{10}Cu_2$ [1120.07]	Chocolate brown	23	>300	55.57 (55.76)	3.92 (3.6)	15.4 (15.01)	–	11.30 (11.35)
(12)	$H_2L^2 + CuCl_2 \cdot 2H_2O + LiOH$	$[(L^2)_2Cu_2Cl_2] \cdot H_2O \cdot EtOH$ $C_{28}H_{26}N_6O_6Cl_2Cu_2$ [740.55]	Dark brown	58	>300	45.59 (45.41)	3.55 (3.54)	11.1 (11.35)	9.3 (9.57)	17.0 (17.16)
(13)	$H_2L^2 + CuBr_2$	$[(L^2)_2Cu_2Br_2]$ $C_{26}H_{18}N_6O_4Br_2Cu_2$ [765.37]	Dark brown	53	>300	40.83 (40.80)	2.29 (2.37)	10.7 (10.98)	–	16.4 (16.61)
(14)	$H_2L^2 + Cu(OAc)_2 \cdot H_2O + 8-HQ$	$[(L^2)_2Cu_2(8-HQ)_2(EtOH)_2]$ $C_{48}H_{42}N_8O_8Cu_2$ [986.01]	Chocolate brown	79	>300	58.39 (58.47)	4.1 (4.29)	11.1 (11.36)	–	12.7 (12.89)
(15)	$H_2L^2 + Cu(OAc)_2 \cdot H_2O + Phen$	$[(L^2)_2Cu_2(OAc)_2(Phen)_2] \cdot 3H_2O$ $C_{54}H_{46}N_{10}O_{11}Cu_2$ [1138.12]	Brown	40	>300	57.06 (56.99)	3.8 (4.07)	12.0 (12.31)	–	10.9 (11.17)
(16)	$H_2L^2 + Cu(OAc)_2 \cdot H_2O + Bac$	$[(L^2)_2Cu_2(H_2O)_2(Bac)_2] \cdot 3H_2O$ $C_{46}H_{46}N_6O_{13}Cu_2$ [1018.00]	Reddish brown	64	>300	54.24 (54.27)	4.3 (4.55)	8.0 (8.26)	–	12.3 (12.48)

^a The yield is calculated on the basis of ligands.

BDH or Merck products. Organic solvents (ethanol, absolute ethanol, methanol, isopropanol, diethylether, dimethylformamide (DMF) and dimethylsulfoxide (DMSO)) were reagent grade chemicals and were used without further purification.

Measurements

Elemental analyses (C, H, N, S and Cl) were carried out at the Microanalytical Center, Cairo University, Giza, Egypt. Analysis of the copper(II) ion followed the dissolution of the solid complex in concentrated HNO_3 , neutralizing the diluted aqueous solutions with ammonia and titrating the metal solutions with EDTA. Melting points of the ligands and their complexes were determined using a Stuart SMP3 melting point apparatus. IR spectra were recorded using KBr discs on FT IR Nicolet IS10 spectrometer. The electronic spectra were recorded at room temperature on a Jasco model V-550 UV/Vis spectrophotometer as Nujol mulls and/or solutions in DMF. ^1H NMR spectra were recorded using a Mercury-300BB (300 MHz). Dimethylsulfoxide, DMSO-d_6 , was used as a solvent and tetramethylsilane (TMS) as an internal reference. ESR spectra of the complexes were recorded at Elexsys, E500, Bruker company. The magnetic field was calibrated with a 2,2'-diphenyl-1-picrylhydrazyl (DPPH) sample purchased from Aldrich. Mass spectra were recorded on GC-2010 Shimadzu Gas chromatography instrument mass spectrometer. Samples were introduced directly to the probe, and the fragmentations were carried out at 300 °C and 70 eV. The magnetic susceptibility measurements were carried out at room temperature using a magnetic susceptibility balance of the type Johnson Matthey, Alfa product, Model No. (MKI). Effective magnetic moments were calculated and corrected using Pascal's constants for the diamagnetism of all atoms in the compounds [33]. Molar conductivities of 10^{-3} M solutions of the solid complexes in DMF were measured on the Corning conductivity meter NY 14831 model 441. TGA-measurements were carried out from room temperature up to 800 °C at a heating rate of 10 °C/min on a Shimadzu-50 thermal analyzer. The biological activity of the

ligands and their metal complexes was studied using the disc diffusion method [34].

Synthesis of the hydrazone ligands

The hydrazone ligands were prepared in two steps. The first step was the formation of 4,6-diacetylresorcinol (DAR) by acetylation of resorcinol [16]. The second step was the condensation of 4,6-diacetylresorcinol stoichiometrically in the molar ratio 1:2 with 3-hydrazino-5,6-diphenyl-1,2,4-triazine or isatin monohydrazone to give the hydrazone ligands: 1-(5-(1-(2-(5,6-diphenyl-1,2,4-triazin-3-yl)hydrazono)ethyl)-2,4-dihydroxyphenyl)ethanone (H_2L^1) and 3,3'-(1,1'-(4,6-dihydroxy-1,3-phenylene)bis(ethan-1-yl-1-ylidene))bis(hydrazine-2,1-diylidene)diindoline-2-one (H_2L^2), respectively (Structure 1). The use of 1:2 or 1:1 (DAR:hydrazino triazine) molar ratios yielded the same product (1:1) not the desired bis compound. The preparation method is as follows:

A solution of 4,6-diacetylresorcinol (0.5 g, 2.58 mmol) in absolute ethanol or methanol (30 mL) was added to 3-hydrazino-5,6-diphenyl-1,2,4-triazine (0.678 g, 2.58 mmol or 1.358 g, 5.16 mmol) in absolute ethanol or isatin monohydrazone (0.831 g, 5.16 mmol) in methanol (30 mL). The reaction mixture was heated under reflux for 8 h which yielded brown products. The precipitate was filtered off, washed several times with absolute ethanol or methanol then with diethylether and finally air-dried. The H_2L^1 and H_2L^2 ligands were recrystallized from acetic acid and methanol–DMF, respectively. The crystals were dried in a desiccator over anhydrous calcium chloride. The analytical and physical data for the ligands and their metal complexes are listed in Table 1.

Synthesis of the metal complexes

The metal salt and the ligand, both dissolved in ethanol, were mixed in the molar ratio 1:1 in case of H_2L^1 ligand and 2:1 (M:L) in case of H_2L^2 ligand and heated under reflux for 8 h. The resulting precipitates were filtered, washed with ethanol then ether and finally air-dried. The complexes were kept in a desiccator over

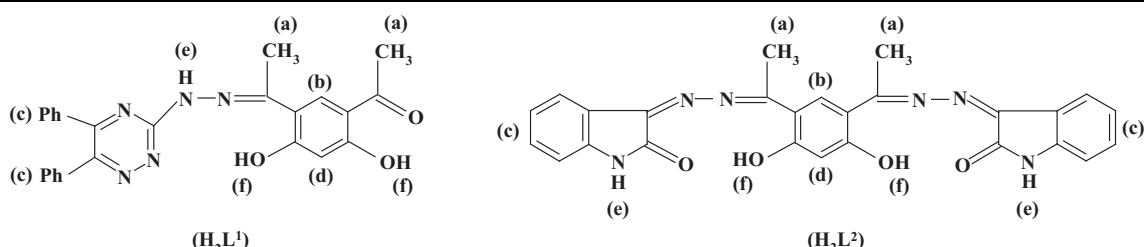
Table 2
Characteristic IR spectral data of the hydrazone, H_2L^1 and H_2L^2 , ligands and their complexes.

No.	Complex	IR spectra (cm^{-1})							
		$\nu(\text{OH})$ and/or $\nu(\text{NH})$ triazine or isatin	$\nu(\text{C}=\text{N})$ azomethine	$\nu(\text{C}=\text{N})$ triazine	$\nu(\text{N}=\text{N})$ triazine	$\nu(\text{C}=\text{O})$ isatin	$\nu(\text{M}-\text{O})$	$\nu(\text{M}-\text{N})$	Other bands
1	H_2L^1 [(HL^1)Cu(OAc)(H_2O)]·1.5 H_2O	3429, 3306	1609	1533	1442	–	–	–	1553 $\nu_{\text{as}}(\text{COO}^-)$, 1484 $\nu_{\text{s}}(\text{COO}^-)$; (bidentate OAc^-)
		3425, 3254	1593	1505	1428	–	579	474	
2	[(HL^1)Cu(H_2O)] NO_3	3446, 3185	1595	1517	1433	–	494	421	1446; $\nu(\text{NO}_3^-)$ (ionic)
3	[(H_2L^1)Cu(SO_4)(H_2O) $_2$]·1.5 H_2O	3467	1558	1507	1430	–	495	445	1098, 1048; $\nu(\text{SO}_4^{2-})$ (monodentate)
4	[(HL^1)CuCl]·1.5 H_2O	3446, 3188	1599	1517	1432	–	570	418	
5	[(HL^1)Cu(H_2O) $_3$]Br· H_2O	3447, 3187	1595	1516	1427	–	579	427	
6	[(HL^1)Cu(8-HQ)(H_2O)]	3341, 3292	1601	1505	1430	–	513	446	1486, $\nu(\text{C}=\text{N})$ 8-HQ
7	[(HL^1)Cu(OAc)(Phen)]	3440	1592	1506	1427	–	531	419	1562 $\nu_{\text{as}}(\text{COO}^-)$, 1445 $\nu_{\text{s}}(\text{COO}^-)$; (monodentate OAc^-), 1540, $\nu(\text{C}=\text{N})$ Phen
8	[(HL^1)Cu(H_2O)(Bac)]·2 H_2O	3577	1589	1505	1423	–	494	462	1685, $\nu(\text{C}=\text{O})$ Bac
9	H_2L^2 [(L^2) $_2$ Cu $_2$ (OAc) $_2$ (EtOH) $_2$]·0.5 H_2O ·0.5EtOH	3420, 3229	1621	–	–	1716	–	–	1548 $\nu_{\text{as}}(\text{COO}^-)$, 1486 $\nu_{\text{s}}(\text{COO}^-)$; (bidentate OAc^-)
		3417	1592	–	–	1710	546	469	
10	[(L^2) $_2$ Cu $_2$]·EtOH	3402, 3217	1594	–	–	1710	577	469	
11	[(L^2) $_2$ Cu $_2$]·2 H_2O	3402, 3217	1594	–	–	1710	577	469	
12	[(L^2)Cu $_2$ Cl $_2$]· H_2O ·EtOH	3442, 3340	1586	–	–	1709	550	459	
13	[(L^2)Cu $_2$ Br $_2$]	3435	1593	–	–	1693	546	491	
14	[(L^2)Cu $_2$ (8-HQ) $_2$ (EtOH) $_2$]	3417	1598	–	–	1710	519	492	1497, $\nu(\text{C}=\text{N})$ 8-HQ
15	[(L^2)Cu $_2$ (OAc) $_2$ (Phen) $_2$]·3 H_2O	3416	1586	–	–	1695	545	490	1551 $\nu_{\text{as}}(\text{COO}^-)$, 1428 $\nu_{\text{s}}(\text{COO}^-)$; (monodentate OAc^-), 1520, $\nu(\text{C}=\text{N})$ Phen
16	[(L^2)Cu $_2$ (H_2O) $_2$ (Bac) $_2$]·3 H_2O	3440	1589	–	–	1710	510	469	1670, $\nu(\text{C}=\text{O})$ Bac

Table 3Electronic spectra, magnetic moments and molar conductivity data of the hydrazone, H₂L¹ and H₂L², ligands and their complexes.

No.	Complex	Electronic spectral bands ^a (nm) λ _{max} ^a (nm)/(ε _{max})	μ _{eff.} ^b B.M.	μ _{compl.} ^c B.M.	Conductance ^d (Ω ⁻¹ cm ² mol ⁻¹)
	H ₂ L ¹	281 (0.28), 331 (0.26), 371 (0.14)	–	–	–
1	[(HL ¹)Cu(OAc)(H ₂ O)]·1.5H ₂ O	474 ^e , 537 ^e sh	2.12	–	8.3
2	[(HL ¹)Cu(H ₂ O)]NO ₃	543 ^d	1.86	–	107.4
3	[(H ₂ L ¹)Cu(SO ₄)(H ₂ O) ₂]·1.5H ₂ O	500 ^e sh, 550 ^e sh	2.15	–	39
4	[(HL ¹)CuCl]·1.5H ₂ O	538 ^d	1.71	–	34.2
5	[(HL ¹)Cu(H ₂ O) ₃]Br·H ₂ O	519 ^d sh, 566 ^d	1.8	–	84.9
6	[(HL ¹)Cu(8-HQ)(H ₂ O)]	520 ^d sh, 578 ^d sh	1.31	–	7.2
7	[(HL ¹)Cu(OAc)(Phen)]	457 ^d , 542 ^d	1.83	–	4.5
8	[(HL ¹)Cu(H ₂ O)(Bac)]·2H ₂ O	486 ^e , 567 ^d	2.1	–	5.2
	H ₂ L ²	266 (0.3), 342 (0.2), 422 (0.18)	–	–	–
9	[(L ²)Cu ₂ (OAc) ₂ (EtOH) ₂]·0.5H ₂ O·0.5EtOH	493 ^d , 564 ^d	1.4	1.66	6.4
10	[(L ²) ₂ Cu ₂]·EtOH	530 ^d , 600 ^d	2.1	2.57	11
11	[(L ²) ₂ Cu ₂]·2H ₂ O	530 ^d , 600 ^d	2.02	2.6	11
12	[(L ²)Cu ₂ Cl ₂]·H ₂ O·EtOH	589 ^d	1.9	2.54	40.3
13	[(L ²)Cu ₂ Br ₂]·H ₂ O	515 ^d	1.62	2.07	24.5
14	[(L ²)Cu ₂ (8-HQ) ₂ (EtOH) ₂]·H ₂ O	519 ^d , 565 ^d	2.01	2.67	10
15	[(L ²)Cu ₂ (OAc) ₂ (Phen) ₂]·3H ₂ O	515 ^d , 558 ^d	2.1	2.74	12.5
16	[(L ²)Cu ₂ (H ₂ O) ₂ (Bac) ₂]·3H ₂ O	474 ^d sh, 573 ^d	2.12	2.8	6.2

^a Solutions in DMF (10⁻³ M), values of ε_{max} are in parentheses and multiplied by 10⁻⁴ (L mol⁻¹ cm⁻¹).^b μ_{eff.} is the magnetic moment of one cationic species in the complex.^c μ_{compl.} is the total magnetic moments of all cations in the complex.^d Nujol mull.^e Concentrated solutions.**Table 4**¹H NMR spectral data of the hydrazone ligands.

						
	H ^a	H ^b	H ^c _{aromatic}	H ^d	H ^e	H ^f
H ₂ L ¹	2.57, 2.66 (6H)	6.38 (1H)	7.34–7.54 (10H)	8.08 (1H)	11.62 (1H; exchangeable with D ₂ O)	12.58, 14.32 (2H; exchangeable with D ₂ O)
H ₂ L ²	2.58, 2.66 (6H)	6.39 (1H)	6.42–8.18 (8H)	8.25 (1H)	10.77 (2H; exchangeable with D ₂ O)	12.61, 13.54 (2H; exchangeable with D ₂ O)

anhydrous calcium chloride. The following detailed preparations are given as examples and the other complexes were obtained similarly.

Synthesis of [(HL¹)Cu(OAc)(H₂O)]·1.5H₂O, complex (1)

About 0.182 g (0.91 mmol) of copper(II) acetate dissolved in 30 mL ethanol was added gradually to 0.4 g (0.91 mmol) of the ligand, H₂L¹, dissolved in 30 mL ethanol. The reaction mixture was heated under reflux for 8 h which resulted a brown precipitate that was filtered off, washed several times with ethanol, diethylether and finally air-dried. The yield was 0.46 g (83%).

Synthesis of [(HL¹)Cu(8-HQ)(H₂O)], complex (6)

About 0.182 g (0.91 mmol) of copper(II) acetate dissolved in 30 mL ethanol was added gradually to 0.4 g (0.91 mmol) of the ligand, H₂L¹, dissolved in 30 mL ethanol. The reaction mixture was heated under reflux for 30 min. and then 0.132 g (0.91 mmol) of 8-hydroxyquinoline (8-HQ) dissolved in ethanol was added to the above mixture. The resulting mixture was heated under reflux for 8 h which resulted a chocolate brown precipitate that was

filtered off, washed several times with ethanol, diethylether and finally air-dried. The yield was 0.58 g (96%).

Antimicrobial activity

The standardized disc-agar diffusion method [34] was followed to determine the activity of the synthesized compounds against the sensitive organisms *Staphylococcus aureus* (ATCC 25923) and *Bacillus subtilis* (ATCC 6635) as Gram positive bacteria, *Salmonella typhimurium* (ATCC 14028) and *Escherichia coli* (ATCC 25922) as Gram negative bacteria and *Candida albicans* (ATCC 10231) and *Aspergillus fumigatus* as fungus strain. The antibiotic chloramphenicol was used as reference in the case of Gram-positive bacteria, cephalothin in the case of Gram-negative bacteria and cycloheximide in the case of fungi.

The compounds were dissolved in DMF which has no inhibition activity to get concentrations of 100 μg mL⁻¹ and 50 μg mL⁻¹. The test was performed on medium potato dextrose agars (PDA) which contain infusion of 200 g potatoes, 6 g dextrose and 15 g agar [35]. Uniform size filter paper disks (3 disks per compound) were