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Template synthesis, characterization and antimicrobial activity of some new complexes with isonicotinoyl hydrazone ligands

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Abstract: Complexes of Cu(II), Ni(II), Co(II) with the 9-anthraldehyde isonicotinoyl hydrazone ligand (HL¹) and the 3,5-di-*tert*-butyl-4-hydroxybenzaldehyde isonicotinoyl hydrazone ligand (H₂L²) were synthesized by the template method. The complexes were characterized by analytical analysis, IR, UV-Vis and ESR spectroscopy, magnetic measurements, conductometry and thermal analysis and the two ligands by ¹H-NMR spectroscopy. From the elemental analysis, 1:2 (metal:ligand) stoichiometry for the complexes of Cu(II), Ni(II) with the ligands HL¹ and H₂L² and 1:1 (metal:ligand) stoichiometry for the complex of Co(II) with the ligand HL¹ are proposed. The molar conductance data showed that the complexes are non-electrolytes. The magnetic susceptibility results coupled with the electronic and ESR spectra suggested a distorted octahedral geometry for the complexes Ni(II)/HL¹, Ni(II)/H₂L² and Cu(II)/H₂L², a tetrahedral stereochemistry for the complex Cu/HL¹ and a square-planar geometry for the complex Co/HL¹. The IR spectra demonstrated the bidentate coordination of the ligands HL¹ and H₂L² by the O=C amide oxygen and the azomethine nitrogen, as well as monodentate coordination of the ligand HL¹ by the azomethine nitrogen in the Cu(II) complex. The antibacterial activity of the ligands and their metallic complexes were tested against *Staphylococcus aureus*, *Escherichia coli* and *Klebsiella pneumoniae*.

Keywords: Cu(II), Ni(II) and Co(II) complexes; template synthesis; isonicotinoylhydrazone; characterization; antibacterial activity.

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INTRODUCTION

Hydrazone ligands and their complexes with different transition metal ions have been thoroughly studied due to their biological activity.¹⁻³ The aroylhydrazones contain in their structure the $-\text{CO}-\text{NH}-\text{N}=\text{C}<$ group that imparts on these chelating agents fungicidal,⁴ antibacterial,⁵ antiparasital⁶ and anticancerous⁷ properties. The complexes of Ni(II), Mn(II) with 6-methylpyridine 2-carboxaldehyde isonicotinoyl hydrazone exhibit antituberculosis activity.⁸ The parameters of the chemical structure, and the physical and electronic characteristics of the ligands are determining factors in the manifestation of bioactivity. In continuation of ongoing studies⁹ on complexes with ligands of the isonicotinoylhydrazone class, the synthesis and study of new mixed-ligand complexes of Cu(II), Ni(II) and Co(II) with such ligands are presented in this paper.

EXPERIMENTAL

All the employed reagents and solvents were of AR grade and were used without further purification. The M and Cl contents were obtained by literature methods¹⁰ and C, H and N were determined with a Hewlett Packard 185 CHN-analyzer. The molar conductivity of the complexes was measured with a HACH-sens ion 5-conductivity meter using 10^{-3} M solutions in DMF. The IR spectra were recorded between $4000-400\text{ cm}^{-1}$ on a BIORAD-FT-IR 135 FTS spectrophotometer using the KBr disc technique. The electronic reflectance spectra (300–1100 nm) were obtained on a VSU-2P Zeiss-Jena spectrophotometer using MgO as the standard. The ESR spectra for the Cu(II) complexes were registered at room temperature (293 K) on a microcrystalline powder with an ART5 spectrophotometer. The magnetic moments were determined by the Faraday method at the room temperature. The ^1H - and ^{13}C -NMR spectra were recorded on a Varian Gemini 300BB instrument in DMSO- d_6 . The thermal analysis was realized with an MOM-Q-1500D derivatograph in air at a heating rate of $5\text{ }^\circ\text{C min}^{-1}$.

Synthesis

A solution of 0.0010 mol of isonicotinoylhydrazine and 0.0010 mol of aldehyde (9-anthraldehyde or 3,5-di-*tert*-butyl-4-hydroxybenzaldehyde) in 75 ml of methanol was refluxed for 5 h on a water bath. Subsequently, reaction mixture was concentrated. The precipitate was filtered and recrystallized from ethanol.

A methanolic solution of anhydrous metal chloride MCl_2 ($\text{M} = \text{Cu(II)}, \text{Co(II)}$ or Ni(II)) (0.0010 mol in 30 ml of MeOH) was added to a mixture of isonicotinoylhydrazine (0.0020 mol in 30 ml of MeOH) and aldehyde (9-anthraldehyde or 3,5-di-*tert*-butyl-4-hydroxybenzaldehyde, 0.0020 mol in 75 ml of MeOH). The reaction mixture was refluxed on a water-bath for 4 h after which a part of the solvent was removed by distillation. The precipitated complexes were filtered off, washed with MeOH and then with diethyl ether and finally dried under vacuum over anhydrous CaCl_2 .

Antibacterial activity

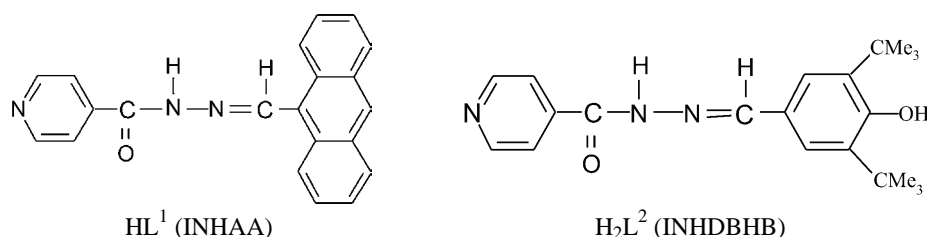
The metal complexes and the free isonicotinoylhydrazone ligands were tested for their activity against the pathogenic strains of bacteria: *Staphylococcus aureus* (Gram-(+)), *Escherichia coli* (Gram-(-)) and *Klebsiella pneumoniae* (Gram-(-)). The paper disc diffusion method¹¹ was applied using DMF as the solvent (the concentration was $125\text{ }\mu\text{g ml}^{-1}$).

RESULTS AND DISCUSSION

The obtained complexes were coloured powders, stable for a long time in the open atmosphere, insoluble in methanol, ethanol, chloroform and acetone. The complexes Co(II)/HL¹, Cu(II)/H₂L² and Ni(II)/H₂L² were soluble in DMF but Cu(II)/HL¹ and Ni(II)/HL¹ were only partly soluble in this solvent. The elemental analysis showed a stoichiometry of 1:2 (metal:ligand) for the complexes, except for the Co(II)/HL¹ complex which had a 1:1 ratio. The analytical data of the ligands and complexes are given in Table I. The presence of lattice water was confirmed by TG analysis. The low values of the molar conductivity supported a non-electrolyte nature for the metal complexes. The structural formulas corresponding to the ligands HL¹ (INHAA) and H₂L² (INHDBHB) are presented in Fig. 1.

TABLE I. Analytical and physical data of the complexes

Compound	M.p. °C	Colour	Found (Calcd.), %					μ_{eff} μ_{B}	A_{m}^{a} $\Omega^{-1} \text{cm}^2 \text{mol}^{-1}$
			M	C	H	N	Cl		
Ligand HL ¹ C ₂₁ H ₁₅ N ₃ O	271	Yellow	–	77.31 (77.53)	4.40 (4.61)	12.73 (12.92)	–	–	–
[Cu(HL ¹) ₂ Cl ₂]	254	Khaki	7.88 (8.09)	64.05 (64.24)	3.64 (3.82)	10.48 (10.70)	8.83 (9.04)	2.16	–
[Ni(HL ¹) ₂ Cl ₂]	272	Dark ochre	7.33 (7.52)	64.42 (64.63)	3.61 (3.84)	10.56 (10.77)	8.89 (9.10)	3.31	–
[Co(HL ¹)Cl ₂]	265	Light ochre	12.74 (12.95)	55.18 (55.39)	3.08 (3.29)	9.04 (9.23)	15.38 (15.60)	2.18	10.47
Ligand H ₂ L ² C ₂₁ H ₂₇ N ₃ O ₂	263	Light yellow	–	71.19 (71.38)	7.43 (7.64)	11.67 (11.89)	–	–	–
[Cu(H ₂ L ²) ₂ Cl ₂]·2H ₂ O	248	Green	7.06 (7.24)	57.28 (57.49)	6.38 (6.61)	9.37 (9.58)	7.88 (8.10)	1.85	11.52
[Ni(H ₂ L ²) ₂ Cl ₂]·4H ₂ O	242	Pale yellow	6.27 (6.46)	55.29 (55.52)	6.61 (6.83)	9.03 (9.25)	7.61 (7.82)	3.29	13.64

^a10⁻³ M solution in DMFFig. 1. The structural formulas of the ligands HL¹ and H₂L².¹H-NMR spectroscopy

Ligand HL¹. ¹H-NMR (300 MHz, DMSO-*d*₆, δ / ppm): 12.32 (1H, *s*, N₈-H), 9.68 (1H, *s*, C₁₀-H), 8.76 (2H, *d*, $J = 10.3$ Hz, C_{2,6}-H), 7.93 (2H, *d*, $J = 11.4$ Hz, C_{3,5}-H), 7.53–7.74 (8H, *m*, anthracene rings).

Ligand H₂L². ¹H-NMR (300 MHz, DMSO-*d*₆, δ / ppm): 11.93 (1H, *s*, N₈-H), 8.83 (2H, *d*, *J* = 6.2 Hz, C_{2,6}-H), 8.40 (1H, *s*, C₁₀-H), 7.85 (2H, *d*, *J* = 6.8 Hz, C_{3,5}-H), 1.41 (18H, *s*, C_{13,15}-*t*-Bu).

The signal of the azomethine proton (-N=CH-) at δ 9.68 or 8.40 ppm in the ¹H-NMR spectrum, as well as the peak at δ 148.38 or 149.31 ppm, assigned to the azomethine carbon in the ¹³C-NMR spectrum, sustain the formation of the isonicotinoylhydrazone ligands.

IR spectroscopy

The characteristic frequencies for ligands HL¹, H₂L² and their complexes are presented in Table II. In the IR spectrum of the 9-anthraldehyde isonicotinoyl hydrazone ligand (HL¹), the amide I band, ν (C=O), was present at 1656 cm⁻¹,¹² and the absorption at 1597 cm⁻¹ is ascribed to the ν (C=N) vibration, which is specific for the azomethine group.¹³ The low intensity band at 602 cm⁻¹ corresponds to the in-plane “ β ” deformation vibration for the pyridine ring.¹⁴ The amide I valence vibration shifted to lower values in the Ni(II) and Co(II) complexes but it exhibited a positive shift in the Cu(II) complex, which support the coordination of only the O=C amide oxygen to the Ni(II) and Co(II) ions.¹⁵ The absorption of the ν (C=N) azomethine group for all the complexes was situated at lower wave numbers than the value for the free ligand, consequently confirming the coordination of the azomethine nitrogen atom to the Cu(II), Ni(II) and Co(II) ions.¹⁶ The “ β ” deformation vibration corresponding to the pyridine ring did not suffer significant shifts in the spectra of complexes, suggesting that the nitrogen from the pyridine ring does not participate in the coordination.¹⁷ The coordination of the azomethine nitrogen of the HL¹ ligand to the Cu(II), Ni(II) and Co(II) ions was also proved by the ν (M-N) vibrations appearing in the range 485–419 cm⁻¹,¹⁸ which were absent in the spectrum of the ligand. The IR spectrum of the 3,5-di-*tert*-butyl-4-hydroxybenzaldehyde isonicotinoyl hydrazone ligand (H₂L²) had an absorption band at 3401 cm⁻¹, attributed to the ν (OH) valence vibration in the phenolic group. The amide I ν (C=O) vibration was situated at 1644 cm⁻¹ and the band at 1593 cm⁻¹ corresponds to the ν (C=N) vibration in the azomethine

TABLE II. Characteristic IR bands (cm⁻¹) for ligands HL¹, H₂L² and their complexes

Compound	ν (NH)	ν (C=O) Amide I	ν (C=N)	δ (NH) Amide II	γ (NH) Amide III	β ring pyridine in plane	ν (M-N)
INHAA (HL ¹)	3046	1656	1597	1552	1365	602	–
[Cu(HL ¹) ₂ Cl ₂]	3049	1663	1572	1554	1372	603	481
[Ni(HL ¹) ₂ Cl ₂]	3051	1641	1568	1517	1378	598	485
[Co(HL ¹)Cl ₂]	3050	1642	1583	1540	1367	601	419
INHDBHB (H ₂ L ²)	3019	1644	1593	1553	1359	590	–
[Cu(H ₂ L ²) ₂ Cl ₂].2H ₂ O	3043	1630	1580	1518	1366	575	498
[Ni(H ₂ L ²) ₂ Cl ₂].4H ₂ O	3041	1607	1575	1550	1364	585	476

group. The weak band at 590 cm^{-1} represents the in-plane “ β ” deformation vibration for the pyridine ring. In the spectra of the Cu(II) and Ni(II) complexes with the H_2L^2 ligand, the intense band at 3260 or 3205 cm^{-1} is attributed to the lattice water present in these complexes.¹⁹ The $\nu(\text{OH})$ valence vibration of the phenolic group appears in the spectra of the Cu(II) and Ni(II) complexes at 3392 and 3386 cm^{-1} , respectively, which reveals that the phenolic group is not deprotonated and is not coordinated to the metal ions. The amide I band and the azomethine vibration band suffered negative shifts in the spectra of the Cu(II) and Ni(II) complexes, suggesting the H_2L^2 ligand coordinated to the Cu(II) and Ni(II) ions *via* the $\text{O}=\text{C}$ amide oxygen and azomethine nitrogen atoms. The band of the “ β ” deformation mode of the pyridine ring suffered shifts to lower values in comparison with that from the free ligand, meaning that the pyridine nitrogen did not participate in the coordination. The coordination of the H_2L^2 ligand to the Cu(II) and Ni(II) ions by the azomethine nitrogen is also supported by the $\nu(\text{M}-\text{N})$ vibration, appearing at 498 or 476 cm^{-1} , which was not found in the spectrum of the ligand.

Electronic and ESR spectra

In its electronic spectrum, the INHAA (HL^1) ligand presents an intense band at 25575 cm^{-1} , which can be assigned to a $n \rightarrow \pi^*$ transition. In the spectra of the complexes, this transition shifts to lower values, with $\Delta\nu$ 1766 – 884 cm^{-1} , indicating coordination of the ligand to the metal ions. The electronic spectrum of the INHDBHB (H_2L^2) ligand presents an absorption maximum at 27624 cm^{-1} due to a $n \rightarrow \pi^*$ transition in the $\text{C}=\text{O}$ and $\text{C}=\text{N}$ chromophoric groups. This transition was found in the spectra of the complexes but shifted to lower frequencies, indicating coordination of the ligand to the metal ions. Information regarding the geometry of the complexes was obtained from the electronic spectra and from the values of the magnetic moments.

The $[\text{Cu}(\text{HL}^1)_2\text{Cl}_2]$ complex presents in its reflectance spectrum a wide band at 12820 cm^{-1} characteristic of the ${}^2\text{T}_2 \rightarrow {}^2\text{E}$ transition, being specific for a Cu(II) ion in a tetrahedral environment. The value of the magnetic moment of $2.16\ \mu_{\text{B}}$ supports the proposed geometry.²⁰

The electronic spectrum of the $[\text{Ni}(\text{HL}^1)_2\text{Cl}_2]$ complex contains two absorption bands at 15948 and 10288 cm^{-1} , corresponding to the ${}^3\text{A}_{2\text{g}} \rightarrow {}^3\text{T}_{1\text{g}}(\text{F})$ (ν_2) and ${}^3\text{A}_{2\text{g}} \rightarrow {}^3\text{T}_{2\text{g}}(\text{F})$ (ν_1) transitions, respectively. These transitions suggest an octahedral stereochemistry for the Ni(II) ion, which is in accordance with the magnetic moment value of $3.31\ \mu_{\text{B}}$.²¹ The ratio ν_2/ν_1 is 1.55 , which is within the range for octahedral complexes of Ni(II). For this complex, the following parameters 10 Dq , B and β were obtained by calculation: $10\text{ Dq} = 10280\text{ cm}^{-1}$, $B = 679\text{ cm}^{-1}$, $\beta = 0.65$. The obtained value of the β parameter indicates a moderate to intense covalent character for the metal–ligands bonds.

In the reflectance spectrum of the $[\text{Co}(\text{HL}^1)\text{Cl}_2]$ complex, the two absorption bands at 21052 cm^{-1} and 16129 cm^{-1} were assigned to the ${}^2\text{A}_{1\text{g}} \rightarrow {}^2\text{E}_{\text{g}}$ and ${}^2\text{A}_{1\text{g}}$

→ ${}^2B_{2g}$, respectively. They are characteristic for square-planar stereochemistry of the Co(II) ion. The magnetic moment of $2.18 \mu_B$ supports this geometry for the low-spin Co(II).²²

The electronic spectrum of the $[Cu(H_2L^2)_2Cl_2] \cdot 2H_2O$ complex contains a wide band situated at approximately 15350 cm^{-1} . This band is characteristic for a Cu(II) ion with tetragonally distorted octahedral stereochemistry and can be assigned to the ${}^2E_g \rightarrow {}^2T_{2g}$ transition.²³ The magnetic moment of $1.85 \mu_B$ suggests a monomeric octahedral geometry.

For the $[Ni(H_2L^2)_2Cl_2] \cdot 4H_2O$ complex, two absorption bands at 15873 and 10256 cm^{-1} were registered in the electronic spectrum. These bands can be assigned to the ${}^3A_{2g} \rightarrow {}^3T_{1g}(F)$ (ν_2) and ${}^3A_{2g} \rightarrow {}^3T_{2g}(F)$ (ν_1) transitions and are specific for the Ni(II) ion in octahedral coordination, which was also confirmed by the magnetic moment of $3.29 \mu_B$. The ratio ν_2/ν_1 is 1.54 , which is within the range found for Ni(II) octahedral complexes. The absorption band associated with the ${}^3A_{2g} \rightarrow {}^3T_{1g}(P)$ (ν_3) transition is covered by the much higher intensity $n \rightarrow \pi^*$ transition characteristic of the ligand. For this complex, the following $10 Dq$, B and β parameters were determined: $10 Dq = 10256 \text{ cm}^{-1}$, $B = 671 \text{ cm}^{-1}$, $\beta = 0.66$. The value of the β parameter indicates a moderate to intense covalent character for the metal–ligands bonds.

The ESR spectrum of a polycrystalline sample of the $[Cu(HL^1)_2Cl_2]$ complex was recorded at room temperature. This complex exhibited a broad and intense slightly anisotropic signal, assigned to the Cu(II) ion in a slightly distorted tetrahedral environment.²⁴ The extremely high intensity of the ESR signal indicates a monomeric structure for the complex.

The ESR spectrum of the $[Cu(H_2L^2)_2Cl_2] \cdot 2H_2O$ complex was recorded on microcrystalline powder at room temperature. The high intensity of the signal confirms the monomeric molecular formula. An axial signal with two “ g ” values at 293 K ($g_{\parallel} = 2.1914$; $g_{\perp} = 2.0685$) was registered. The anisotropic shape of the spectrum with $g_{\parallel} > g_{\perp}$ indicates a compound with an axially distorted octahedral geometry. The g_{\parallel} and g_{\perp} values were > 2 , corresponding to an axial symmetry with all main axes disposed parallelly. The fact that $g_{\parallel} > g_{\perp} > 2.0023$ supports a ground state of the Cu(II) ion with the unpaired electron in the $d_{x^2-y^2}$ orbital and from the ESR spectrum there results an octahedral stereochemistry tetragonally distorted by elongation.²⁵ The G parameter determined with the formula $G = ((g_{\parallel} - 2)/(g_{\perp} - 2))$ was less than 4 and, consequently, there is considerable interaction in the solid state in this complex.

Thermal analysis

The thermal decomposition of the synthesized complexes was studied in the air in the range $25\text{--}700 \text{ }^\circ\text{C}$ and the results are listed in Table III.

For the Cu(II) and Ni(II) complexes with the H₂L² ligand, lattice water was lost between 80–170 °C. All complexes underwent decomposition at high temperatures ($t > 245$ °C) and the HL¹ and H₂L² ligands were eliminated in two stages. The stable residue at the final temperature (≈ 700 °C) contained CuO, NiO or Co₃O₄. The results obtained from the thermal analysis supported the molecular formulas assigned to the complexes.

The structural formulas assigned to the complexes are presented in Fig. 2.

TABLE III. Thermal analysis data of the prepared complexes

Complex	Total mass losses, %		Temperature, °C	Loss, %
	Theoretical	Experimental		
[Cu(HL ¹) ₂ Cl ₂]	89.80	89.54	250–385	39.68
			385–670	49.86
			670–700	9.89 exp./res. 10.13 calc./res.
[Ni(HL ¹) ₂ Cl ₂]	87.77	88.64	80–240	–
			240–365	49.23
			365–495	39.41
			495–700	9.72 exp./res. 9.58 calc./res.
[Co(HL ¹)Cl ₂]	82.29	82.40	257–405	67.12
			405–675	15.28
			675–700	17.42 exp./res. 17.64 calc./res.
[Cu(H ₂ L ²) ₂ Cl ₂].2H ₂ O	90.92	91.08	80–170	3.81
			170–375	32.33
			375–625	54.94
			625–700	8.91 exp./res. 9.07 calc./res.
[Ni(H ₂ L ²) ₂ Cl ₂].4H ₂ O	91.73	91.15	85–160	7.72
			160–370	65.93
			370–525	17.50
			525–700	8.06 exp./res.
				8.23 calc./res.

Antibacterial activity

The antibacterial activity of the HL¹, H₂L² ligands and of their complexes were studied on the Gram-(+) bacteria *Staphylococcus aureus* and on the Gram-(–) bacteria *Escherichia coli* and *Klebsiella pneumoniae*. The experimental data expressed as the diameter of the inhibition zone (in mm) of bacterial growth by the tested compounds are presented in Table IV.

The results show that HL¹ and H₂L² exhibited weak activity. The HL¹ ligand was slightly more active than the H₂L² ligand. Against the three bacterial agents, the Cu(II)/HL¹ complex was more active than the HL¹ ligand, while the Ni(II)/HL¹ and Co(II)/HL¹ complexes showed lower activity. The Cu(II)/H₂L² and

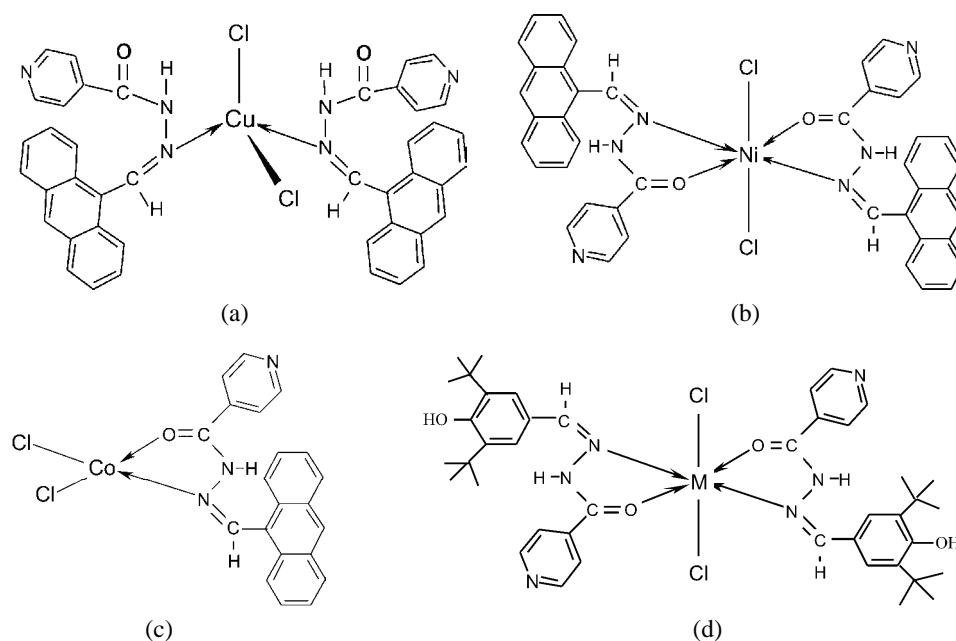


Fig. 2. The structural formulas of the complexes: a) $[\text{Cu}(\text{HL}^1)_2\text{Cl}_2]$, b) $[\text{Ni}(\text{HL}^1)_2\text{Cl}_2]$, c) $[\text{Co}(\text{HL}^1)\text{Cl}_2]$ and d) $[\text{M}(\text{H}_2\text{L}^2)_2\text{Cl}_2] \cdot x\text{H}_2\text{O}$ ($\text{M} = \text{Cu}(\text{II}): x = 2$; $\text{M} = \text{Ni}(\text{II}): x = 4$).

TABLE IV. Antibacterial activity data for 0.125 mg bacterium ml^{-1}

Compound	Inhibition zone of bacterial growth, mm		
	<i>Staphylococcus aureus</i>	<i>Escherichia coli</i>	<i>Klebsiella pneumoniae</i>
INHAA (HL^1)	6	4	5
$[\text{Cu}(\text{HL}^1)_2\text{Cl}_2]$	9	6	7
$[\text{Ni}(\text{HL}^1)_2\text{Cl}_2]$	2	3	2
$[\text{Co}(\text{HL}^1)\text{Cl}_2]$	3	2	2
INHDBHB (H_2L^2)	3	2	2
$[\text{Cu}(\text{H}_2\text{L}^2)_2\text{Cl}_2] \cdot 2\text{H}_2\text{O}$	5	6	4
$[\text{Ni}(\text{H}_2\text{L}^2)_2\text{Cl}_2] \cdot 4\text{H}_2\text{O}$	7	8	6

$\text{Ni}(\text{II})/\text{H}_2\text{L}^2$ complexes possessed a higher antibacterial activity than the free H_2L^2 ligand. The data in Table IV show that *S. aureus* was much more inhibited by the $\text{Cu}(\text{II})/\text{HL}^1$ complex, giving the opportunity for its use in medical practice. The increased activity of the chelates can be explained based on the overtone concept and the Tweedy chelation theory.²⁶ According to the overtone concept of cell permeability, the lipid membrane surrounding the cell favours the passage of only lipid-soluble materials, which means that liposolubility is an important factor controlling antimicrobial activity. On chelation, the polarity of a metal ion is greatly reduced due to overlap with the ligand orbital and the partial sharing of its

positive charge with the donor groups. In addition, it is also due to delocalization of the π -electrons over the whole chelate ring, thus enhancing the penetration of the complexes into the lipid membranes and the blocking of the metal binding sites of the enzymes of the micro-organisms.

CONCLUSIONS

The Cu(II), Ni(II) and Co(II) complexes with 9-anthraldehyde isonicotinoyl hydrazone (HL¹), as well as those of Cu(II) and Ni(II) with 3,5-di-*tert*-butyl-4-hydroxybenzaldehyde isonicotinoyl hydrazone (H₂L²) are described. They were synthesized by the template method and characterized by their analytical and spectral data. A tetrahedral geometry was assigned to the Cu(II)/HL¹ complex, a square-planar to the Co(II)/HL¹ complex and octahedral one to the Ni(II)/HL¹, Cu(II)/H₂L² and Ni(II)/H₂L² complexes. The Cu(II)/HL¹, Cu(II)/H₂L² and Ni(II)/H₂L² complexes demonstrated higher antibacterial activity than the corresponding free ligands.

ИЗВОД

ТЕМПЛАТНА СИНТЕЗА, КАРАКТЕРИЗАЦИЈА И АНТИМИКРОБНА АКТИВНОСТ НОВИХ КОМПЛЕКСА СА ИЗОНИКОТИНОИЛ-ХИДРАЗОНСКИМ ЛИГАНДИМА

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Темплатном методом синтетисани су комплекси Cu(II), Ni(II), и Co(II) са 9-антралдеhid-изоникотиноил-хидразонским (HL¹), као и 3,5'-ди-*терц*-бутил-4-хидроксибензалдеhid-изоникотиноил-хидразонским лигандом (H₂L²). Комплекси су окарактерисани аналитичким подацима, IR, UV-Vis, ESR спектрима, магнетним мерењима, кондуктометријом, термичком анализом и за два лиганда ¹H-NMR спектрима. Из елементалне анализе стехиометрија за комплексе Cu(II), Ni(II) са лигандима HL¹ и H₂L² је 1:2 (метал:лиганд), док је за комплекс Co(II) са HL¹ лигандом предложена 1:1 (метал:лиганд). Подаци моларне проводљивости указују да су комплекси неелектролити. Магнетна суспектибилност са електронским и ESR спектрима сугерише дисторговану октаедарску геометрију за комплексе Ni(II)/HL¹, Ni(II)/H₂L², Cu(II)/H₂L², тетраедарску стереохемију за комплекс Cu/HL¹ и квадратно-планарну геометрију за комплекс Co/HL¹. IR спектри показују бидентатну координацију лиганда HL¹, H₂L² преко O=C амидног кисеоника и азометинског азота, као и монодентатну координацију лиганда HL¹ помоћу азометинског азота у комплексу Cu(II). Тестирана је и нађена антибактеријска активност лиганда и њихових металних комплекса према *Staphylococcus aureus*, *Escherichia coli* и *Klebsiella pneumoniae*.

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