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BİR DİAZENİL LİGAND: 4-[(DİSİYANOMETİL)]BENZOİKASİT: İLE METAL ASETAT KOMPLEKSLERİNİN SPEKTROSKOPİK KARAKTERİZASYONU VE ANTİBAKTERİYAL AKTİVİTESİ

COMPLEXES OF METAL ACETATE WITH A DIAZENYL LIGAND: 4-[(DICYANOMETHYL)]BENZOIC ACID: SPECTROSCOPIC CHARACTERIZATION AND ANTIBACTERIAL ACTIVITY

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Özet

3-Aminopropiltrimetoksisilan ve hidroksibenzaldehitin reaksiyonu sonucu diazenil grubu içeren 4-[(disiyanometil)diazenil]benzoikasit sentezlenerek yeni Pb(II), UO_2(II), Cu(II), Ni(II), Co(II) ve Zn(II) kompleksleri sentezlendi. Ligand ve komplekslerin yapıları elemental analiz, UV-vis., IR, ^{1}H-NMR, ^{13}C-NMR, TGA, DTA ve magnetik ölçümler kullanılarak belirlendi. Co(II) ve Ni(II) kompleksler paramagnetik olup oktahedral, Zn(II) kompleksi diamagnetik olup oktahedral, Cu(II) kompleksi paramagnetic olup kare düzlem, UO_2(II) ve Pb(II) kompleksler diamagnetic olup karedüzlem yapıya sahiptirler. Tüm komplekslerin antibakteriyel aktiviteleri *Escherichia coli* ATCC-25922, *Enterococcus faecalis* ATCC 29212, *Staphylococcus aureus* ATCC 25923 ve *Salmonella typhimurium* CCM 583'e karşı testedildi. Test sonuçları değerlendirildi. Bu sonuçlardan sadece Ni(II) and Zn(II) komplekslerinin *Staphylococcus aureus'*un büyümesini inhibe ettiği görülmüştür.

Anahtar Kelimeler:*Spektroskopi, diazenil, asetat tuzları, termogravimetrik analiz, kare düzlem,antibacterial aktivite*

Abstract

The new Pb(II), UO_2(II), Cu(II), Ni(II), Co(II) and Zn(II) complexes were synthesized with the ligand containing the diazenyl group 4-[(dicyanomethyl)diazenyl]benzoic acid (L) which were prepared by

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the reaction of hydroxybenzaldehyde with 3-aminopropyltrimethoxysilane. The structure of the ligand and its complexes were determined by using elemental analysis, UV-visible, IR, ^{1}H-NMR, ^{13}C-NMR, TGA, DTA and magnetic measurements. The Co(II) and Ni(II) complexes are paramagnetic with octahedral structure; the Zn(II) complex is diamagnetic with octahedral structure; the Cu(II) complex is paramagnetic and have square planar structure; UO_2(II) and Pb(II) complexes are diamagnetic and have square planar structure. All of the complexes were tested for their antibacterial activities against *Escherichia coli* ATCC-25922, *Enterococcus faecalis* ATCC 29212, *Staphylococcus aureus* ATCC 25923 and *Salmonella typhimurium* CCM 583. The test results were also evaluated. It was found that only Ni(II) and Zn(II) complexes are able to inhibit the growth of *Staphylococcus aureus.*

Keywords: *Spectroscopy, diazenyl; acetate salts; thermo gravimetric analysis; square planar; antibacterial activities.*

1. INTRODUCTION

The coordination chemistry of azoderivatives of β-diketones is still a little explored area of research, notwithstanding their interesting chemical and structural features. Azo compounds (diazenes) also represent an important group of organic compounds, which found versatile applications, for example, as azo dyes, as reagents inorganic synthesis, in the complexation of metal ions and in biological medical studies, plastics, lasers, liquid crystalline displays, electro optical devices and ink-jet printers in high-technology areas [1-5].

Studies have also shown that diazo compounds exhibit properties similar to that of Schiff bases [6].

In this article, we report the complex formation and characterization using different techniques, in particular, elemental analyses, magnetic measurements, infrared and electronic spectra and thermal studies. The thermal decomposition of the complexes have been studied and indicates that not only the crystallization and coordinated water are lost but also that the decomposition of the ligand from the complexes is necessary to interpret the successive mass losses. Finally, the complexes synthesized were tested for their antibacterial activities against *Escherichia coli* ATCC-25922, *Enterococcus faecalis* ATCC 29212, *Staphylococcus aureus* ATCC 25923 and *Salmonella typhimurium* CCM 583. The ligand didn't showed antibacterial activity against these bacteria while compare with ampicillin (10µg, Oxoid).

2. EXPERIMENTAL

All reagents were reagent grade materials purchased from Merck or Aldrich Chemical Co. Ltd., and were used without further purification.

Elemental analyses (C, H, N) were carried out on a Leco CHNS-O model 932 elemental analyzer. ^{1}H-NMR spectra were recorded using a model Bruker GmbH DPX-400 MHz FT spectrometer in DMSO-d_6 at room temperature. IR spectra were recorded Perkin Elmer Precisely Spectrum One spectrometer on KBr discs in the wave number range of 4000-400 cm^{-1}. Electronic spectral studies were conducted on a Shimadzu model UV-1700 Spectrophotometer in the wavelength 1000-200 nm. Magnetic susceptibilities measurements were performed using the standard Gouy tube technique using $Hg[Co(SCN)_4]$ as a calibrant. Thermal analyses (TGA and DTA) were carried out in nitrogen atmosphere with a heating rate of 10 °C/min. using Shimadzu DTG-60 AH (Shimadzu DSC 60 A) thermal analyzers.

2.1. Antibacterial Activity Studies

Disk diffusion method was used for antibacterial activity of ligand and its complexes. Antibacterial activity against *Escherichia coli* ATCC-25922, *Enterococcus faecalis* ATCC 29212, *Staphylococcus aureus* ATCC 25923 and *Salmonella typhimurium* CCM 583 were investigated. Mueller–Hinton agar (oxoid) for all bacterial strains was used. The media were melted at 100 °C, autoclaved at 121 °C for 15 min, cooled 45 °C to 50 °C and were poured into plates of 9 cm diameter in quantities of 20 mL, and left on a flat surface to solidify and the surface of media was dried at 37 °C. Then, preparation of the inoculums was used colony growth method in Mueller–Hinton broth to a turbidity equivalent 0.5 Mc Farland (10^8 cfu mL^{-1}). Petri dishes were streaked using sterile cotton swab. The surface of the media was allowed to dry 3–5 min at room temperature. The 10 mg mL^{-1} (in DMSO, E. Merck), compound impregnated blank discs (Oxoid) were applied to the surface of inoculated plates. The Mueller–Hinton agar plates were incubated at 35±2 °C for 18–24 h. The plates were examined and the diameter of the inhibition zone was measured surrounding discs. As the standard antibiotic ampicillin (10 µg, Oxoid) was used [7].

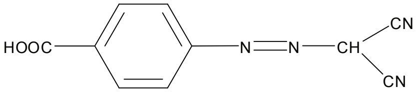


Figure 1. Synthesis of the 4-[(dicyanomethyl)diazenyl]benzoic acid (L)

2.2. Synthesis of the Ligand (L)

4-[(Dicyanomethyl)diazenyl]benzoic acid (L) was synthesized by the method described in the literature [8]. Prepared from 4-amino-benzoic acid; crystallization from acetic acid gave orange yellow crystals (80%), mp: 280 °C; FW: 214.00 g/mol. ^{1}H-NMR: 15.00 (s, 1H, O$\underline{H}$), 7.30-8.00 (m, 4H, Ar-C$\underline{H}$), 3.40 (H, C$\underline{H}$). ^{13}C NMR (DMSO-d_6): 86.26 (CH), 119.30, 122.00, 128.12, 130.20, 154.81, (aryl, 2CN), 170.35 (C=O) ppm. IR(KBr) cm^{-1}:) ν(cm^{-1}): $3428_{(br)}$ (O-H), 3194-3200 (Ar-C-H), 2930-2890 (C-H), 2214,2213 (C≡N), 1580-1570 (N=N), 1594 (C=C); *Anal.* Calcd for $C_{10}H_6N_4O_2$: C, 56.08; H, 2.80; N, 26.42. Found: C, 56.11; H, 2.10; N, 26.11.

2.3. Synthesis of the Metal Complexes

The preparation of complexes was carried out by mixing methanol solution (100 mL) of metal acetate (10 mmol) and a hot DMF solution of the ligand (10 mmol, 2.14 g) in 1:1 molar ratio. The resulting solution was refluxed for 6 h, precipitation of the products occurred after cooling and were filtered off, washed several times with methanol diethyl ether and air-dried. All of the other complexes were prepared similarly. The complexes were obtained with 68-80% yields.

$[CoL(H_2O)_3(AcO)_2].H_2O$ complex: Yield: (72 %). mp: 366 °C. FW: 462.73 g/mol. Color: Dark brownish. IR(KBr) cm^{-1}:) ν(cm^{-1}): 3500-3450 (O-H), 3054 (Ar-C-H), 2923-2846 (C-H), 2214 (C≡N), 1565 (N=N), 1593 (C=C), 535 (M-O), 478 (M-N); *Anal.* Calcd for $C_{14}H_{20}N_4O_{10}Co$: C, 36.31; H, 4.32; N, 12.10. Found: C, 36.26; H, 3.98; N, 12.09.

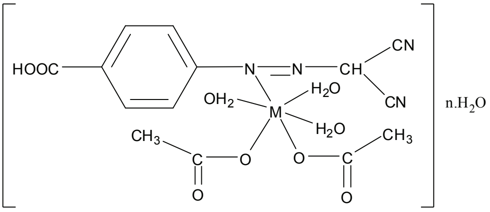
$[NiL(H_2O)_3(AcO)_2].3H_2O$ complex: Yield: (70 %). mp: 360 °C. FW: 498.71 g/mol. Color: Orange Yellow. IR(KBr) cm^{-1}:) ν(cm^{-1}): 3428-3236 (O-H), $3170_{(br)}$ (Ar-C-H), 2933 (C-H), 2212 (C≡N), 1555 (N=N), 1596 (C=C), 510 (M-O), 488 (M-N); *Anal.* Calcd for $C_{14}H_{24}N_4O_{12}Ni$: C, 33.69; H, 4.81; N, 11.23. Found: C, 33.40; H, 4.80; N, 10.81.

$[CuL(H_2O)(AcO)_2].2H_2O$ complex: Yield: (67 %). mp: 335 °C. FW: 449.86 g/mol. Color: Black. IR(KBr) cm^{-1}:) ν(cm^{-1}): 3435-3346 (O-H), 3280 (Ar-C-H), 2994,2928 (C-H), 2214 (C≡N), 1544 (N=N), 1591 (C=C), 503 (M-O), 466 (M-N); *Anal.* Calcd for $C_{14}H_{18}N_4O_9Cu$: C, 37.38; H, 4.03; N, 12.45. Found: C, 37.35; H, 3.91; N, 12.42.

$[ZnL(H_2O)_3(AcO)_2].3H_2O$ complex: Yield: (68 %). mp: 340 °C. FW: 505.37 g/mol. Color: Pale yellow. IR(KBr) cm^{-1}:) ν(cm^{-1}): 3436 (O-H), 3260 (Ar-C-H), 2923,2846 (C-H), 2214 (C≡N), 1569 (N=N), 1592 (C=C), 507 (M-O), 493,480 (M-N); *Anal.* Calcd for $C_{14}H_{24}N_4O_{12}Zn$: C, 33.25; H, 4.78; N, 11.08. Found: C, 33.13; H, 4.84; N, 11.05.

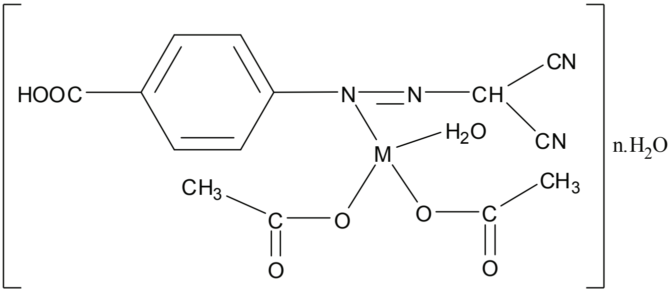
$[PbL(H_2O)(AcO)_2]$ complex: Yield: (75 %). mp: 350 °C. FW: 557.48 g/mol. Color: Green. IR(KBr) cm^{-1}:) ν(cm^{-1}): 3430-3315 (O-H), 3234 (Ar-C-H), 2933,2856 (C-H), 2214 (C≡N), 1567 (N=N), 1594 (C=C), 523 (M-O), 482 (M-N); *Anal.* Calcd for $C_{14}H_{14}N_4O_7Pb$: C, 30.16; H, 2.53; N, 10.05. Found: C, 30.23; H, 2.54; N, 10.10.

$[UO_2L(H_2O)(AcO)_2]$ complex: Yield: (70 %). mp: 345 °C. FW: 620.31 g/mol. Color: White. IR(KBr) cm^{-1}:) ν(cm^{-1}): 3428-3336 (O-H), 3225 (Ar-C-H), 2923 (C-H), 2213 (C≡N), 1558 (N=N), 1599 (C=C), 518 (M-O), 466 (M-N); *Anal.* Calcd for $C_{14}H_{14}N_4O_7UO_2$: C, 27.11; H, 2.27; N, 9.03. Found: C, 28.93; H, 2.30; N, 9.10.



M	Co(II)	Ni(II)	Zn(II)
n	1	3	3

Figure 2. Suggested structure of the octehedral Co(II), Ni(II) and Zn(II) complexes of the ligand



M	Cu(II)	Pb(II)	UO_2(II)
n	2	-	-

Figure 3. Suggested structure of the square-planar Cu(II), Pb(II) and UO_2(II) complexes of the ligand

3. RESULTS AND DISCUSSION

All the complexes are insoluble in EtOH and MeOH but are soluble in DMSO and DMF to give stable solutions at room temperature.

The elemental analyses data of the Pb(II), UO_2(II), Cu(II), Ni(II), Co(II) and Zn(II) complexes of the ligand (L) containing the diazenyl group are given in the experimental section. The experimental elemental analyses results of the complexes are in good compliance with the theoretical calculations. The complexes were characterized by the following physical studies.

3.1. Spectral Studies

The N=N stretching frequencies for the ligand occurring at approximately 1580-1570 cm^{-1} are shifted to lower frequencies (1569–1544 cm^{-1}) in the complexes, indicating the coordination of the diazo nitrogen to metal [9].

The IR spectra of ligand showed absorption bands corresponding to the COOH group such as 3300-3236 (br, OH, COOH). The bands located at 1670 cm^{-1} and the deformation vibrations in the region between 780 and 500 $cm^{-1}$are assigned to the stretching vibration of carboxylic acid group in ligand [10-12]. This band remains unchanged in the complexes coordination compounds indicating the non-participation of carboxylic acid oxygen towards coordination.

Aromatic C-H and aliphatic C-H bands occurred at 3280-3054 cm^{-1} and 2994-2846 cm^{-1}, respectively. Conclusive evidence of the banding is also shown by the observation that new bands in the spectra of the coordination compounds appear 493–466 at and 535-503 cm^{-1} range assigned to [(M-O) and (M-N)] bands stretching vibrations which are not observed in the spectra of the ligand [13,14]. The IR bands of the cyanide groups are observed for ligand at 2214 and 2212 $cm^{–1}$ [15].

In the investigated complexes, the bands observed in the region 3280–3428, 842–863 and 700–720 $cm^{–1}$ are attributed to OH stretching, rocking and wagging vibrations, respectively due to the presence of water molecules [16-18]. The presence of hydrate water was also established and supported by TG/DTA analysis of these complexes.

The coordination of an acetate group in all the complexes is indicated by the appearance of two new bands in the regions 1660–1618 and 1326–1382 $cm^{–1}$ assignable to υ_{as}(COO) and υ_{s}(COO) modes, respectively. The value of $\Delta\upsilon = \upsilon_{as}(COO)-\upsilon_{s}(COO) = 278–292\ cm^{–1}$ suggests the unidentate coordination of the acetate group [19]. In the uranyl complex two additional sharp bands are observed at 920 and 861 $cm^{–1}$, which are assigned to $\upsilon_{as}(UO_2)$ and $\upsilon_{s}(UO_2)$ modes, respectively. This observation suggests that the O=U=O moiety is virtually linear in this complex [20].

All the metal complexes show broad to medium bands in the region 3550–3000 cm^{-1}, which are assignable to OH stretching vibrations due to the presence of free COOH groups and/or due to the presence water molecules [21].

3.2. Electronic Spectra and Magnetic Studies

The measured magnetic moments for the synthesized metal complexes are given in the Table 1. The electronic spectra of the metal complexes $[CoL(H_2O)_3(AcO)_2].H_2O$, $[NiL(H_2O)_3(AcO)_2].3H_2O$, $[CuL(H_2O)(AcO)_2].2H_2O$, $[ZnL(H_2O)_3(AcO)_2].3H_2O$, $[PbL(H_2O)(AcO)_2]$ and $[UO_2L(H_2O)(AcO)_2]$ were measured in DMF at room temperature, and the wave lengths of maximum absorbance are reported in the experimental section. All the metal complexes show an intense band at ca. 261–265 nm, which is assigned to $\pi \rightarrow \pi^*$ transitions of the benzene ring [22]. The band around 305–399 nm in the UV visible spectra of the metal complexes is due to the $n \rightarrow \pi^*$ transition of the nonbonding electrons present on the nitrogen of the diazenyl group [23]. The spectra of all the metal complexes show an intense band at ca. 400–460 nm, which can be assigned to charge transfer transition.

The electronic spectra of Co(II) complex showed two spin-allowed transitions at 612 nm and 460 nm assignable to $^4T_{1g}(F) \rightarrow {}^4A_{2g}(F)$ and $^4T_{1g}(F) \rightarrow {}^4T_{1g}(P)$ transitions, respectively, that are in conformity with octahedral arrangements for Co(II) ion. The magnetic moment of Co(II) complex is 4.88 B.M., which suggests the high spin six coordinated octahedral arrangement of ligand molecules around the metal ion [24,25,13].

The electronic spectra of Ni(II) complex showed the bands at 680 nm 377 nm to the $^3A_{2g} \rightarrow {}^3T_{1g}(F)$ and $^3A_{2g} \rightarrow {}^3T_{1g}(P)$ transitions, respectively, which suggests octahedral geometry. The magnetic moment value (2.95 B.M.) is further confirmation [26,27].

The electronic spectra of Cu(II) complex showed two bands at 650 nm 400, 420 nm, assignable to the transition $^2B_{1g} \rightarrow {}^2E_{1g}$ and $^2B_{1g} \rightarrow {}^2A_{1g}$ and intra-ligand charge transfer bands. These data and the magnetic moment value of 1.72 B.M. suggest square-planar geometry around Cu(II) [28].

The electronic spectra of UO_2(II) displays three spectral bands at 265, 342 and 480 nm. These are assigned to $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ and $^1\sum_g + {}^3\pi_u$ transitions [29], respectively. The last one of the transition is typical of the O=U=O symmetric stretching frequency of the first excited state [13,30].

The spectral data of the Zn(II) complex indicates that it is an octahedral complex of the type $[ZnL(H_2O)_3(AcO)_2].3H_2O$ [31,32].

The Pb(II) and UO_2(II) complexes were found to be diamagnetic as expected.

Table 1. Electronic spectral data of the ligand (L) and its complexes (nm) (in DMF) and magnetic moments.

Compounds	Wavelength in nm (ε)			μ_{eff} (B.M.)
Ligand (L) $C_{10}H_6N_4O_2$	261-324 (4000-3913) $\pi \rightarrow \pi^*$, $n \rightarrow \pi^*$			
$[CoL(H_2O)_3(AcO)_2].H_2O$	312- 387(592, 1064) $\pi \rightarrow \pi^*$	460 (1130) $^4T_{1g}(F) \rightarrow {}^4T_{1g}$ (P)	612 (42) $^4T_{1g}(F) \rightarrow {}^4A_{2g}$ (F)	4.88
$[NiL(H_2O)_3(AcO)_2].3H_2O$	305 (627,) $\pi \rightarrow \pi^*$, $n \rightarrow \pi^*$	377(263) $^3A_{2g} \rightarrow {}^3T_{1g}(P)$	687(29) $^3A_{2g} \rightarrow {}^3T_{1g}(F)$	2.95
$[CuL(H_2O)(AcO)_2].2H_2O$	312-378(588,3263) $\pi \rightarrow \pi^*$, $n \rightarrow \pi^*$	400, 420(3374, 3524) $^2B_1 \rightarrow {}^2A_1$	626(99) $^2B_1 \rightarrow {}^2E$	1.72
$[ZnL(H_2O)_3(AcO)_2].3H_2O$	370(934) $\pi \rightarrow \pi^*$, $n \rightarrow \pi^*$	410-418 (897-372) (Charge transfer bands)		Dia

[PbL(H_2O)($AcO)_2$]	305, 362 (627, 2177) $\pi \rightarrow \pi^*$, $n \rightarrow \pi^*$	401, 420, 570 (1456, 1521, 24)	Dia
[UO_2L(H_2O)($AcO)_2$]	304-399 (4000, 3957) $\pi \rightarrow \pi^*$, $n \rightarrow \pi^*$	414-458(3957-4000)	Dia

ε = molar extinction coefficient (L mol^{-1} cm^{-1}).

3.3. Thermal Study

The compounds were studied by thermogravimetric analysis (TGA) and differential thermal analysis (DTA) to evaluate their thermal properties. The ligand is stable up to 70.00°C and its decomposition started at this temperature. The ligand shows five-step weight loss. The loss of the CN and acetato groups and simultaneously in the first and second steps between 72.00-380.50°C with one exothermic DTA peak at 331.25°C The experimental mass loss of 47.69 % agrees well with the calculated mass loss of 47.62 %.

The thermal decomposition studies of Cu(II), Co(II), Ni(II) Pb(II), UO_2(II) and Zn(II) complexes show no appreciable change at about 50 °C when heated. The thermal analytical data of the Cu(II) complex shows a 7.69% loss in weight (theor. 8.00%), Co(II) complex shows a 3.08% loss in weight (theor. 3.89%), Ni(II) complex shows a 10.76% loss in weight (theor. 10.83%) and Zn(II) complex shows a 10.76% loss in weight (theor. 10.68%) indicating the absence of hydration water molecules in these complexes [32].

In the decomposition process of the CoL($H_2O)_3$($AcO)_2$].H_2O complex, the mass losses corresponded to three coordination water and two acetate groups leaving in the second stage of the decomposition 36.93 % (theor. 37.17%). The DTA curve of the Co(II) complex shows one exothermic peak at 270.60°C.

The thermogram of [PbL(H_2O)($AcO)_2$] shows stability to 180.00°C, above which a weight loss by 24.61% (theor. 24.41%) corresponds to removal of two acetate and one water groups. The second stage ending at 300.10-450.00°C is assigned to the elimination of CH-($CN)_2$ groups from the ligand accompanied by a weight loss of 10.77% (theor. 11.67%) [32]. The DTA curve of the Pb(II) complex shows three exothermic peak at 125,00 186.00 and 270.35°C. The exothermic peak is likely due to the remove of acetate and CH-($CN)_2$.

The TGA curve of [CuL(H_2O)($AcO)_2$].2H_2O complex shows a first weight loss of due to elimination of two hydration water molecule at 200°C. It shows a further weight loss of 30.76% (theor. 30.22%) at 200-320.20°C corresponding to removal of two acetates and one water molecule. The subsequent steps correspond to the removal of the organic part of the ligand.

The weight losses for UO_2(II), Ni(II), and Zn(II) complexes were found to be approximately the same, when expressed as the percentages calculated stoichiometrically from their chemical formulas. The decomposition of the all of complex is irreversible.

Single crystals of the complexes could not be isolated from any solutions, thus no definitive structure could be described. However, the analytical, spectroscopic and magnetic data enable us to propose the possible structures.

Table 2. Antibacterial activity

Chemicals / Bacteria	*Escherichia coli* ATCC-25922	*Enterococcus faecalis* ATCC 29212	*Staphylococcus aureus* ATCC 25923	*Salmonella typhimurium* CCM 583
	Zone Diameter (mm)			
$[CoL(H_2O)_3(AcO)_2].H_2O$	0	0	0	0
$[NiL(H_2O)_3(AcO)_2].3H_2O$	0	0	10	0
$[CuL(H_2O)(AcO)_2].2H_2O$	0	0	0	0
$[ZnL(H_2O)_3(AcO)_2].3H_2O$	0	0	11	0
$[PbL(H_2O)(AcO)_2]$	0	0	0	0
$[UO_2L(H_2O)(AcO)_2]$	0	0	0	0
Ampicillin	16	22	32	26

4. CONCLUSIONS

From the satisfactory analytical data, magnetic studies, thermal and various spectral studies it is concluded that the metal: ligand stoichiometric ratio is 1:1 in all the complexes. The overall spectral studies indicate that the ligand behaves in a monobasic bidentate (O,N) manner. The proposed structures of the synthesized complexes are shown in Figures 2 and 3. The Ni(II) and Zn(II) complex showed at middle level antibacterial activity against the test bacterias while compare with ampicillin (10µg, Oxoid). But ligand and other complexes did not exhibit antibacterial activity against the test bacterias. For these reasons, ligand and other complexes may not be used to prepare new active agent. The results are presented in Table 2.

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