

Synthesis, Characterization and Antibacterial Activity of some Cu (II), Hg (II) Complexes with a Mixture of N-(benzothiazol-2-yl) benzamide and Amines or Diphosphines Ligands

Ruba E. Mageed¹, Ahmed A. Irzoqi^{1*}, Sayran Sattar Saleh², Montazer K. Mensoor³

¹ University of Tikrit, College of Education for Pure Science, Department of chemistry, Tikrit, Iraq

² University of Kirkuk, College of Science, Department of chemistry, Kirkuk, Iraq

³ International Center for Training, Research & Development (ICTRD), Baghdad, Iraq

*Corresponding author: ahmedirzoqi@tu.edu.iq

Abstract:

Benzothiazole derivatives play vital role in some biological applications. The important coordinating sites of the Benzothiazole derivatives have participated in the preparation of many complexes. In this study, a number of Cu(II) and Hg(II) complexes of N-(benzothiazol-2-yl)benzamide(**Btba**) containing co-ligands such as Amines (2-aminopyridine, 3-aminopyridine, 4-aminopyridine, Bipy, Phen) or diphosphines (dppm, dppe) have been prepared. The prepared complexes have been characterized by Molar conductivity, magnetic susceptibility, UV-VIS spectra, IR spectra, ³¹P-NMR, ¹H-NMR and ¹³C-NMR spectra. The suggestion of geometric around the metal ions can be tetrahedral form. That the (**Btba**) ligand acted like a bidentate ligand through the N atom of five member ring in benzothiazole and O atom of carbonyl group, while the amines or diphosphines ligands coordinated through the N atoms or P atoms respectively, where the dppm ligand behaves in complex [Hg₂(Btba)₂(dppm)₂]Cl₄ (**9**) as a bridge ligand while the dppe behaves as a bidentate ligand in complex [Hg(Btba)(dppe)]Cl₂ (**10**).

The biological activity of these prepared complexes against four bacterial species was investigated in this study: *Staphylococcus Epidermidis* and *Staphylococcus aureus* (gram positive) and *E. Coli*, *Citrobacter Freundii* (gram negative) (gram negative). As a reference, amikacin was used. The prepared complexes were more effective than amikacin against *Staphylococcus epidermidis*. The (**9**) complex, on the other hand, was more active against *Staphylococcus aureus* than amikacin until at minimum concentration. In the minimum concentration, the (**3,5,6,8, and 10**) complexes were more active against *E. Coli*, and the (**1,3,4,5,7,8,9, and 10**) complexes were more active against *Citrobacterfreundii* than amikacin.

Keywords: Benzothiazole derivatives, Mercury complexes, diphosphines, antibacterial activity

Introduction:

Benzothiazole is one of the bicyclic ring systems that are important in organic preparations as a basic nucleus for other organic (1,2) or inorganic derivatives in the formation of complexes (3,4). Benzothiazole possesses effective N, S donor atoms and has different consistency methods and its

importance increases as a ligand through derivatives that have donor atoms such as N, S, and O (5,6).

The benzothiazole derivatives are well known among the biologically active bicyclic ring systems containing both S and N atoms (7). Many benzothiazoles are used in medicine as anti-cancer (8), anti-inflammatory (9), anti-parasitic (10) and anti-viral agents (11).

Because of their synthetic stability and antibacterial action of their metal complexes, aminobenzothiazole derivatives are very significant (12). Mercury (II) and copper (II) complexes of 6-methyl-2-aminobenzothiazole have previously been shown to have increased antibacterial activity against *Alternaria alternate*, *Aspergillus niger*, *Penicillium fumiculosus* and *Curvularia lunata* (13).

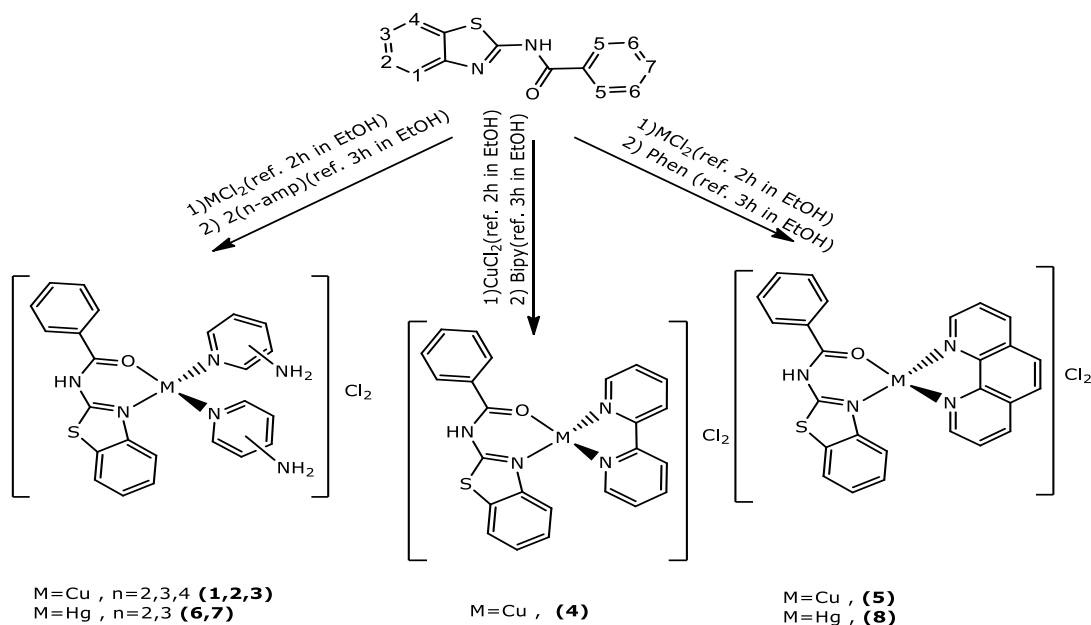
The purpose of the present study is to investigate the synthesis and characterization of specific Cu(II) and Hg(II) complexes with co-ligands such as Amines or Phosphines N-(benzothiazol-2-yl)benzamide (Btba) and the biological activities of a number of the prepared complexes at (0.01, 0.001, 0.0001 mg/ml) concentrations against four *Staphylococcus epidermidis* and *Staphylococcus aureus*.

Experimental Section:

The chemicals were delivered and used without being purified. Melting points were measured using a Stuart (SMP30) melting point apparatus and are provided as uncorrected values. The molar conductivity of the prepared complexes (10^{-3} M) recently dissolved DMSO solution was calculated with a (Starter 3100c) digital conductivity meter. At room temperature, the magnetic susceptibility of the prepared complexes was determined using the Sherwood magnetic susceptibility balance. An Elementar vario El III CHN elemental analyzer was used to document the CHN analysis. A Bruker 400 MHz spectrometer was used to test nuclear magnetic resonance in DMSO- d_6 as a solvent. The IR spectra of complexes with KBr were measured with a Shimadzu FT-IR 8400S spectrophotometer in the 400-4000 cm^{-1} range. PG instruments T80 UV-Vis spectrophotometer was used to test UV-Visible spectra in the spectrum (200-900nm).

Preparation of N-(1,3-benzothiazol-2-yl)benzamide (Btba) ligand (14)

At 50–60°C, a mixture of 2-aminobenzothiazole (0.751 g, 5.0mmol) and triethylamine (0.300 g, 5.0mmol) in dry dioxane (50 mL) was stirred for 30 minutes. By dropwise, a solution of benzoyl chloride (0.702 g, 5.0mmol) in dry dioxane (50 mL) was added. The mixture was stirred for 2 hours before being dumped onto crushed ice. The isolated solid was obtained by filtration and washed with a 1% potassium bicarbonate aqueous solution before being recrystallized in ethanol, yielding yellowish white crystals (Btba): 0.413 g (32 percent). m.p. of 186–188°C: The following values were calculated for $\text{C}_{14}\text{H}_{10}\text{N}_2\text{OS}$ (254.31 g mol^{-1}): C, 66.12; H, 3.96; N, 11.02; and S, 12.61. C, 66.90 percent; H, 3.83 percent; N, 10.65 percent; S, 12.94 percent; IR (KBr): 3230 (NH), 1672 (C=O), 1598 (C=C aromatic), 1554 (C=N), 1444, 1296, 1272, 750 cm^{-1} ; ^1H NMR (400MHz, DMSO- d_6 , (ppm): 12.90 (1H, s, NH); 8.15 (2H, d (Ar-CH).



Scheme 1: The Propose Structure of (1-8) complexes.

Preparation of (1-10) complexes:

A solution of (**Btba**) (0.762, 3.0mmol) in (30ml) absolute ethanol was added to the one mole of CuCl_2 (0.403g, 3.0mmol), or HgCl_2 (0.814g, 3.0mmol) in (30ml) absolute ethanol. Then, the mixture was refluxed for two hours with stirring. Then a solution in (30ml) absolute ethanol of two moles equivalents of one of ligands (2amp, 3amp, 4amp) or one mole equivalents of one of ligands (Bipy, Phen, dppm, dppe) was added to the mixture which was refluxed for three hours with stirring, separated as a precipitate, filtered, and dried in the air.

$[\text{Cu}(\text{Btba})(2\text{amp})_2]\text{Cl}_2$ (**1**) 1.324g, (76%). m.p. 209–212°C: *Anal.* Calc. for $\text{C}_{24}\text{H}_{22}\text{Cl}_2\text{CuN}_6\text{OS}$ (576.99 g mol⁻¹): C, 49.96; H, 3.84; N, 14.57; S, 5.56, Found: C, 49.57; H, 3.48; N, 14.12; S, 5.73%; IR (KBr): 3276 (NH), 3178 (NH₂), 1672 (C=O), 1596 (C=C_{Aromatic}), 1554 (C=N), 1525 (C=N amine), 1442, 1296, 1272, 727 cm⁻¹; $\mu(\text{BM})$: 1.92.; $\Lambda(\text{ohm}^{-1}.\text{cm}^2.\text{mol}^{-1})$: 72.4.

$[\text{Cu}(\text{Btba})(3\text{amp})_2]\text{Cl}_2$ (**2**) 1.417g, (81%). m.p. >236°C (dec.): *Anal.* Calc. for $\text{C}_{24}\text{H}_{22}\text{Cl}_2\text{CuN}_6\text{OS}$ (576.99 g mol⁻¹): C, 49.96; H, 3.84; N, 14.57; S, 5.56, Found: C, 49.63; H, 3.52; N, 14.09; S, 5.45%; IR (KBr): 3240 (NH), 3167 (NH₂), 1671 (C=O), 1596 (C=C_{Aromatic}), 1552 (C=N), 1530 (C=N amine), 1444, 1295, 1274, 729 cm⁻¹; $\mu(\text{BM})$: 1.91.; $\Lambda(\text{ohm}^{-1}.\text{cm}^2.\text{mol}^{-1})$: 69.4.

$[\text{Cu}(\text{Btba})(4\text{amp})_2]\text{Cl}_2$ (**3**) 1.398g, (80%). m.p. 266–269°C: *Anal.* Calc. for $\text{C}_{24}\text{H}_{22}\text{Cl}_2\text{CuN}_6\text{OS}$ (576.99 g mol⁻¹): C, 49.96; H, 3.84; N, 14.57; S, 5.56, Found: C, 49.50; H, 3.61; N, 14.37; S, 5.49%; IR (KBr): 3222 (NH), 3178 (NH₂), 1670 (C=O), 1598

(C=C_{Aromatic}), 1550 (C=N), 1518 (C=N amine), 1446, 1286, 1238, 723 cm⁻¹.; μ (BM): 1.92.; Λ (ohm⁻¹.cm².mol⁻¹):83.5.

[Cu(Btba)(Bipy)]Cl₂(4) 1.267g, (77%). m.p. 214–217°C: Anal. Calc. for C₂₄H₁₈Cl₂CuN₄OS (544.94 g mol⁻¹): C, 52.90; H, 3.33; N, 10.28; S, 5.88, Found: C, 52.67; H, 2.94; N, 9.81; S, 5.53%.; IR (KBr): 3325 (NH), 1679 (C=O), 1593 (C=C_{Aromatic}), 1541 (C=N), 1438 (C=N amine), 1296, 1284, 765, 696 cm⁻¹.; μ (BM): 1.95.; Λ (ohm⁻¹.cm².mol⁻¹):77.3.

[Cu(Btba)(Phen)]Cl₂(5) 1.356g, (79%). m.p. 196–199°C: Anal. Calc. for C₂₆H₁₈Cl₂CuN₄OS (568.96 g mol⁻¹): C, 54.89; H, 3.19; N, 9.85; S, 5.64, Found: C, 54.45; H, 2.77; N, 9.51; S, 5.47%.; IR (KBr): 3396 (NH), 1647 (C=O), 1606 (C=C_{Aromatic}), 1537 (C=N), 1423 (C=N amine), 1340, 1217, 840, 707 cm⁻¹.; μ (BM): 1.93.; Λ (ohm⁻¹.cm².mol⁻¹):76.6.

[Hg(Btba)(2amp)₂]Cl₂(6)1.823g, (85%). m.p. 252–255°C: Anal. Calc. for C₂₄H₂₂Cl₂HgN₆OS (714.03 g mol⁻¹): C, 40.37; H, 3.11; N, 11.77; S, 4.49, Found: C, 39.95; H, 2.73; N, 11.35; S, 4.51%.; IR (KBr): 3350 (NH), 3458 (NH₂), 1664 (C=O), 1618 (C=C_{Aromatic}), 1558 (C=N), 1539 (C=N amine), 1487, 1286, 754 cm⁻¹.¹H NMR (400MHz, DMSO-d₆, δ (ppm): 12.76 (1H, s, NH); 8.27 (2H, d, J=7.68 Hz, H-C6 2amp); 8.17 (2H, d, J=7.45 Hz, H-5); 8.00 (1H, d, J=7.78 Hz, H-1); 7.81 (1H, d, J=7.95 Hz, H-4); 7.72 (2H, t, J=7.82 Hz, H-C4 2amp); 7.66 (1H, t, J=7.32 Hz, H-7); 7.57 (2H, t, J=7.32 Hz, H-6); 7.46 (1H, t, J=7.59 Hz, H-3); 7.37 (2H, t, J=7.99 Hz, H-C5 2amp); 7.30 (1H, t, J=7.54 Hz, H-2); 7.24 (1H, d, J=7.91 Hz, H-C3 2amp); 5.82 (4H, s, 2NH₂);¹³C NMR (400 MHz, DMSO-d₆): δ 161.64 (C=O); 159.41 (C=N); 154.95 (C2-2amp); 149.03 (C-N); 144.92 (C-N); 137.42 (C4-2amp); 136.23 (C-CO); 131.53 (C-S); 130.78 (Ar-CH); 127.62(C6-2amp); 126.57 (Ar-CH); 124.89(C3-2amp); 124.48(Ar-CH); 121.97(C5-2amp); 121.64 (Ar-CH) 114.12 (Ar-CH); 109.75 (Ar-CH); Λ (ohm⁻¹.cm².mol⁻¹):85.4.

[Hg(Btba)(3amp)₂]Cl₂(7)1.572g, (73%). m.p. 237–240°C: Anal. Calc. for C₂₄H₂₂Cl₂HgN₆OS (714.03 g mol⁻¹): C, 40.37; H, 3.11; N, 11.77; S, 4.49, Found: C, 39.87; H, 2.96; N, 11.41; S, 4.37%.; IR (KBr): 3336 (NH), 3434 (NH₂), 1672 (C=O), 1602 (C=C_{Aromatic}), 1546 (C=N), 1521 (C=N amine), 1444, 1282, 748 cm⁻¹.¹H NMR (400MHz, DMSO-d₆, δ (ppm): 12.87 (1H, s, NH); 8.66 (2H, d, ⁴J=5.40 Hz, H-C23amp); 8.34 (2H, d, J=9.35 Hz, H-5); 8.16 (2H, d, J=7.65 Hz, H-C6 3amp); 8.06 (1H, d, J=7.84 Hz, H-1); 7.82 (1H, d, J=8.29 Hz, H-4); 7.70 (2H, t, J=7.10 Hz, H-7); 7.59 (2H, t, J=7.21 Hz, H-6); 7.49 (1H, t, J=7.75 Hz, H-3); 7.37 (1H, t, J=7.81 Hz, H-2); 7.31 (2H, t, J=10.00 Hz, H-C5 3amp); 7.19 (2H, d, J=9.01 Hz, H-C43amp); 5.92 (4H, s, 2NH₂);¹³C NMR (400 MHz, DMSO-d₆): δ 161.55 (C=O); 159.84 (C=N); 153.13 (C-N); 152.43 (C-N); 148.12 (C3-3amp); 138.35 (C6-3amp); 133.19 (C-CO); 131.43 (C-S); 130.38 (Ar-CH); 129.12 (C2-3amp); 128.57 (Ar-CH); 128.29 (C4-3amp); 126.28 (Ar-CH); 123.81 (C5-3amp); 119.51 (Ar-CH) 112.20 (Ar-CH); 109.64 (Ar-CH).; Λ (ohm⁻¹.cm².mol⁻¹):82.1.

[Hg(Btba)(Phen)]Cl₂(8) 1.356g, (79%). m.p. 293–295°C: Anal. Calc. for C₂₆H₁₈Cl₂HgN₄OS (706.01 g mol⁻¹): C, 44.23; H, 2.57; N, 7.94; S, 4.54, Found: C, 44.45; H, 2.34; N, 7.51; S, 4.47%.; IR (KBr): 3323 (NH), 1670 (C=O), 1589 (C=C_{Aromatic}), 1554 (C=N), 1502 (C=N amine), 1417, 1276, 848, 736 cm⁻¹.¹H NMR (400MHz, DMSO-d₆, δ (ppm): 12.91 (1H, s, NH); 9.21 (2H, broad s, H1-

Phen); 8.81 (2H, d, J=7.95 Hz, H3-Phen); 8.20 (2H, s, H4-Phen); 8.15 (2H, d, J=7.25 Hz, H-5); 8.07 (2H, m, H2-Phen); 8.04 (1H, d, J=8.22 Hz, H-1); 7.68 (1H, d, J=8.67 Hz, H-4); 7.68 (1H, t, J=7.34 Hz, H-7); 7.58 (2H, t, J=7.64 Hz, H-6); 7.49 (1H, t, J=7.99 Hz, H-3); 7.36 (1H, t, J=7.98 Hz, H-2); ^{13}C NMR (400 MHz, DMSO-*d*6): δ 162.42 (C=O); 153.32 (C=N); 152.61 (C-N); 149.72 (C-Phen); 145.62 (C-Phen); 135.78 (C-Phen); 133.24 (C-CO); 131.79 (C-S); 130.59 (Ar-CH); 129.59 (Ar-CH); 128.67 (Ar-CH); 128.41 (Ar-CH); 128.15 (C-Phen); 126.72 (Ar-CH); 126.08 (C-Phen); 123.76 (Ar-CH); 122.93 (C-Phen); 119.85 (Ar-CH).; $\Lambda(\text{ohm}^{-1}.\text{cm}^2.\text{mol}^{-1})$:70.6.

$[\text{Hg}_2(\text{Btba})_2(\text{dppm})_2]\text{Cl}_4(9)$ 1.847g, (67%). m.p. $>350^\circ\text{C}$ (dec.): Anal. Calc. for $\text{C}_{78}\text{H}_{64}\text{Cl}_4\text{Hg}_2\text{N}_4\text{O}_2\text{P}_4\text{S}_2$ (1820.39 g mol^{-1}): C, 51.46; H, 3.54; N, 3.08; S, 3.52, Found: C, 50.96; H, 5.28; N, 3.07; S, 3.14%.; IR (KBr): 3247 (NH), 1674 (C=O), 1652 (C=C_{Aromatic}), 1552 (C=N), 1458, 1423, 1276, 1116, 748, 692 cm^{-1} . $^{13}\text{P}\{^1\text{H}\}$ NMR (400MHz, DMSO-*d*6, $\delta(\text{ppm})$: 27.54.; ^1H NMR (400MHz, DMSO-*d*6, $\delta(\text{ppm})$: 12.46 (1H, s, NH); 8.16 (2H, d, J=7.67 Hz, H-5); 8.03 (1H, d, J=7.70 Hz, H-1); 7.80 (1H, d, J=7.86 Hz, H-4); 7.68 (1H, t, J=7.35 Hz, H-7); 7.58 (2H, t, J=7.49 Hz, H-6); 7.48 (8H, m, H-Ar); 7.35 (14H, m, H-Ar); 2.89 (2H, s, CH₂); $\Lambda(\text{ohm}^{-1}.\text{cm}^2.\text{mol}^{-1})$:121.7.

$[\text{Hg}(\text{Btba})(\text{dppe})]\text{Cl}_2(10)$ 1.973g, (71%). m.p. 275–278°C: Anal. Calc. for $\text{C}_{40}\text{H}_{34}\text{Cl}_2\text{HgN}_2\text{OP}_2\text{S}$ (924.22 g mol^{-1}): C, 51.98; H, 3.71; N, 3.03; S, 3.47, Found: C, 51.57; H, 3.41; N, 2.75; S, 3.24%.; IR (KBr): 3286 (NH), 1681 (C=O), 1604 (C=C_{Aromatic}), 1531 (C=N), 1454, 1431, 1242, 1101, 748, 692 cm^{-1} .; $^{13}\text{P}\{^1\text{H}\}$ NMR (400MHz, DMSO-*d*6, $\delta(\text{ppm})$: 36.55.; ^1H NMR (400MHz, DMSO-*d*6, $\delta(\text{ppm})$: 12.89 (1H, s, NH); 8.15 (2H, d, J=5.44 Hz, H-5); 8.04 (1H, d, J=7.19 Hz, H-1); 7.88 (8H, m, H-Ar); 7.61 (16H, m, H-Ar); 7.36 (1H, t, J=7.43 Hz, H-3); 7.18 (1H, t, J=7.47 Hz, H-2); 2.73 (2H, s, CH₂); 2.67 (2H, s, CH₂); $\Lambda(\text{ohm}^{-1}.\text{cm}^2.\text{mol}^{-1})$:74.3.

Results and Discussion:

The methods used to make the mixed ligand complexes are assisted in their tests and have good materials with good purity. They stayed in a wind constantly. It was partially dissolve the complexes in organic solvents such as ethanol or acetone, but soluble in (DMF) or DMSO.

The molar electrical conductivity of the complexes solutions in DMSO at (10^{-3}M) recorded at (25°C) showed the (1-8,10) complexes were ionic by 1:2 negative to positive ion. $[\text{Hg}_2(\text{Btba})_2\mu(\text{dppm})_2]\text{Cl}_4(9)$ complex showed an increase in molar conductivity which supports the current proposed formula of the complex. In addition, these results support the magnetic sensitivity values of Cu complexes as these values were between ($1.91\text{--}1.96 \text{ ohm}^{-1}.\text{cm}^2.\text{mol}^{-1}$) these values are included within the values of the tetrahedral complexes (15,16).

UV-Vis. spectra of the prepared Cu(II) complexes have shown the charge transfer bands within the ($25575\text{--}30581 \text{ cm}^{-1}$) rangewhich are indicated in Table (1). In addition, a d-d transition bands were shown within the ($16529\text{--}15275 \text{ cm}^{-1}$) range and they were not broad enough to explain the other transfers of $^2\text{B}_{2g} \rightarrow ^2\text{B}_{1g}$ and $^2\text{B}_{2g} \rightarrow ^2\text{A}_{1g}$ for octahedral complexes. So, they were interpreted as a $^2\text{T}_2 \rightarrow ^2\text{E}$ transfer of tetrahedral complexes (17).

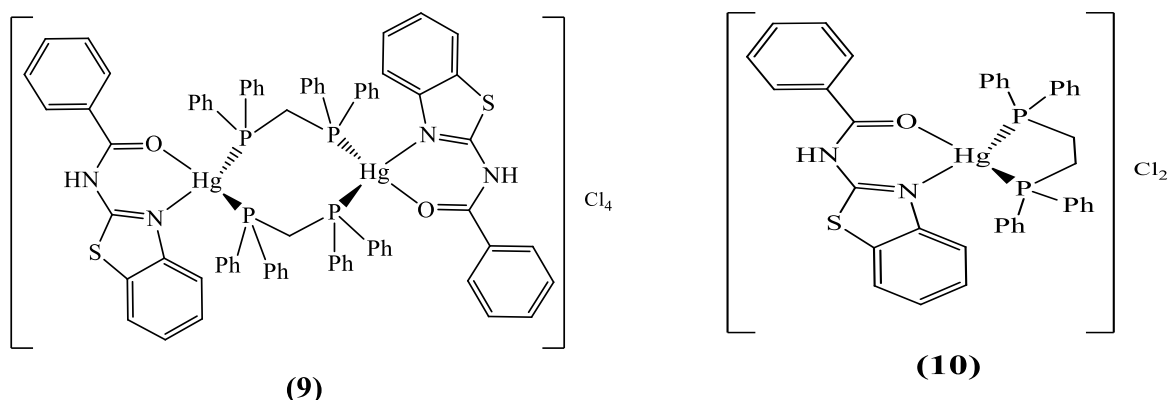
Table 1: Values of electronic transition of Cu (II) complexes

No.	Compounds	Transitions (nm) cm ⁻¹			
		$\pi \rightarrow \pi^*$	$n \rightarrow \pi^*$	C.T.	$^2T_2 \rightarrow ^2E$
	(Btba)	(239) 41841	(267) 37453	-	-
1	[Cu(Btba)(2amp) ₂]Cl ₂	(234) 42735	(264) 37879	(381) 26247	(604) 16556
2	[Cu(Btba)(3amp) ₂]Cl ₂	(236) 42373	(265) 37736	(387) 25840	(605) 16529
2	[Cu(Btba)(4amp) ₂]Cl ₂	(235) 42553	(263) 38023	(391) 25575	(605) 16529
4	[Cu(Btba)(Bipy)]Cl ₂	(236) 42373	(263) 38023	(335) 29851	(640) 15625
5	[Cu(Btba)(Phen)]Cl ₂	(235) 42553	(264) 37879	(337) 29674	(638) 15674

The spectrum of (**Btba**) ligand showed a new band of the carbonyl group $\nu(\text{C=O})$ at (1670 cm⁻¹), while the $\nu(\text{NH})$ group showed at (3240 cm⁻¹) and the $\nu(\text{C=N})$ group displayed at (1552 cm⁻¹).

The spectrum of the complexes showed a carbonyl group within the range of (1646-1681 cm⁻¹). The peaks shifted to lower or higher frequency and that indicates the bonding of O atom of the carbonyl groups with the metal ion. The $\nu(\text{C=N})$ group bands showed a range of (1531-1558 cm⁻¹) and the peaks shifted to lower or higher frequency (18), this illustrates the bonding with the metal. The $\nu(\text{C=N})$ bands of amines in (**1-8**) complexes showed a range of (1423-1539 cm⁻¹), and the $\nu(\text{P-Ar})$ bands of the (**9, 10**) complexes showed at of (1423, 1431 cm⁻¹), the $\nu(\text{P-C})$ bands showed a range of (1101-1116 cm⁻¹) respectively (19-21).

The ¹H-NMR spectra of the complexes (**6-10**) showed the signals of the ligand protons (**Btba**) and it is observed singlet of the proton of NH group within the (12.46-12.91 ppm) range (22). On the other hand, integration ratio in the complexes (**6**) and (**7**) showed coordination of two (2-amp) or (3-amp) molecules against one molecule of (**Btba**) ligand. Spectrum of complex (**8**) has shown signals of phen protons at 9.21 (s, 2H), 8.81 (d, 2H, J = 7.95 Hz), 8.20 (s, 2H), 8.07 (m, 2H), confirming the consistency of one phen molecule versus a (**Btba**) ligand molecule. The ¹H-NMR spectra of (**9, 10**) complexes also showed multiple signals within the δH = (7.88-7.35 ppm) range which belong to the protons of the phenyl rings in dppm and dppe ligands, the signal of CH₂ groups appeared as a singlet at (δH = 2.89 ppm) for (**9**) complex and two singlet at (δH = 2.73, 2.67 ppm) for (**10**) complex, and the integration value indicated the presence of one molecule of phosphine coordinated with the metal versus a molecule of (**Btba**) ligand. The ³¹P-NMR spectrum has shown a single signal with a positive value at (δP = 27.54 ppm). This indicates the consistency of the dppm as a bridging and not as a chelating ligand. This could propose a binuclear dimer formula for complex (**9**), while complex (**10**) showed a positive single signal at (δP = 36.55 ppm), where dppe is believed to behave like a chelating ligand due to its high flexibility compared with dppm (23-25). and as shown in Figure 2.

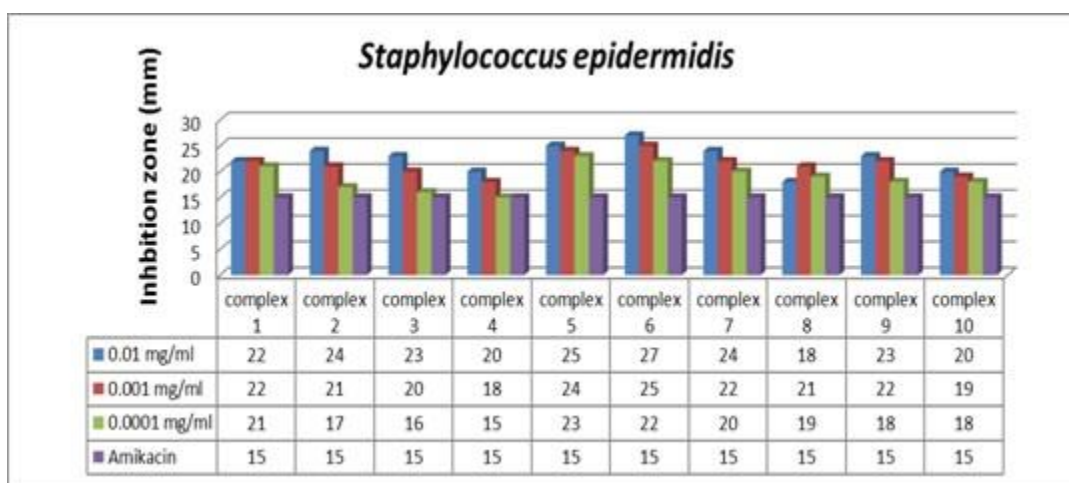


Scheme 2: The propose structure of $[\text{Hg}_2(\text{Btba})_2(\text{dppm})_2]\text{Cl}_4$ (9) and $[\text{Hg}(\text{Btba})(\text{dppe})]\text{Cl}_2$ (10).

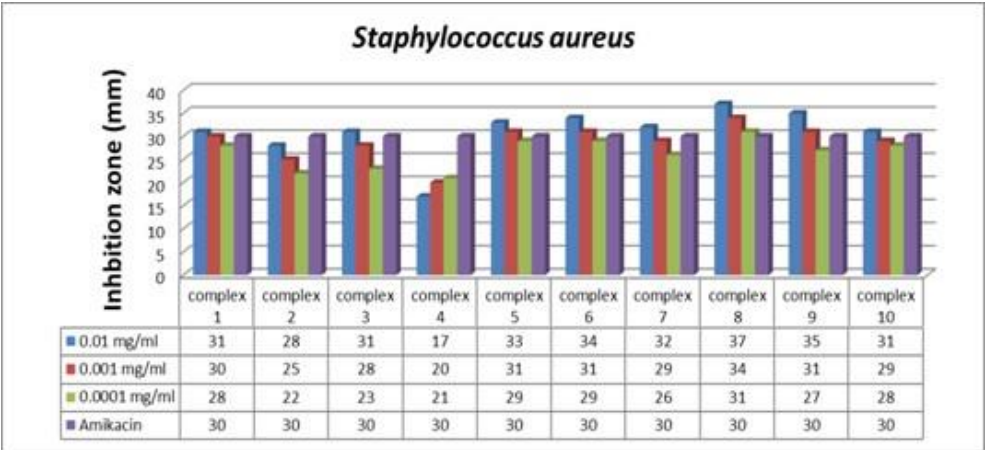
Biological activity of the prepared complexes

The biological activity of the prepared complexes counter to four bacterial species: *Staphylococcus aureus*, *Staphylococcus epidermidis*, *Citrobacterfreundii* and *E.coli*, was tested using the hole method with the antibiotic Amikacin as a reference material. Because of the higher toxicity of Hg(II) complexes, they displayed greater action against bacteria forms than Cu(II) complexes (26).

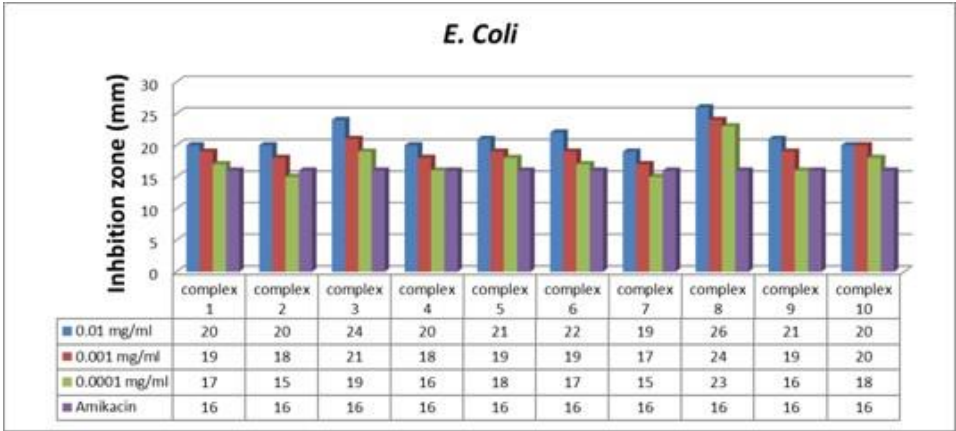
At the minimum inhibition concentration (MIC) all of the prepared complexes (Scheme: 3) were more effective than amikacin against *Staphylococcus epidermidis*, on the other side, the (8) complex outperformed amikacin in terms of action against *Staphylococcus aureus*, the complexes (3,5,6,8, and 10) were more active against *E. Coli* and the complexes (1,3,4,5,7,8,9, and 10) were more active against *Citrobacterfreundii* than amikacin.



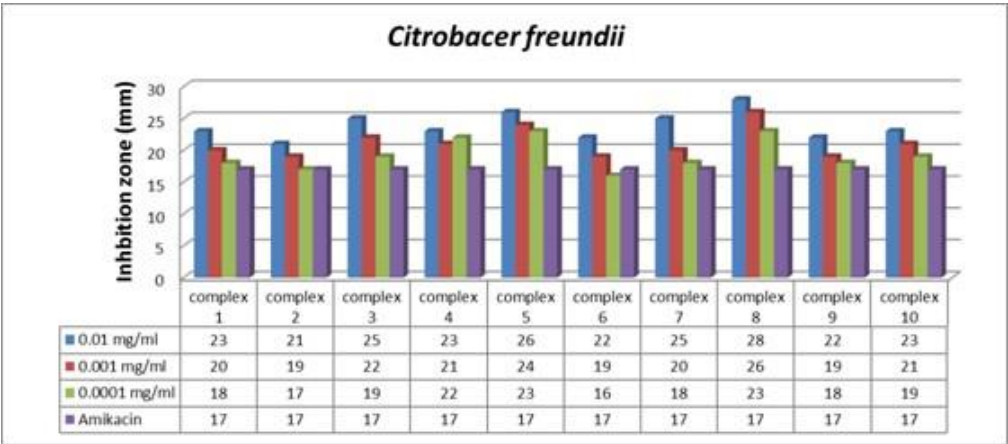
(a)



(b)



(c)



(d)

Scheme 3: The inhibitory activity of prepared complexes in the growth of a number of positive (a), (b) and negative (c), (d) bacteria.

Conclusion:

The prepared complexes (**1-10**) were tetrahedral complexes with formula $[\text{Cu}(\text{Btba})_2\text{Cl}_2]$ and $[\text{M}(\text{Btba})(\text{L})_n]\text{Cl}_2$ were $\text{M}=\text{Cu}$, ($n=2$, $\text{L}=2\text{-aminopyridine}$, 3-aminopyridine , 4-aminopyridine) complexes (**1-3**), ($n=1$, $\text{L}=\text{Bipy}$, Phen) complexes (**4,5**), or $\text{M}=\text{Hg}$ ($n=2$, $\text{L}=2\text{-aminopyridine}$, 3-aminopyridine) complexes (**6,7**), ($n=1$, $\text{L}=\text{Phen}$, dppe) complexes (**8,10**). Excepting the $[\text{Hg}_2(\text{Btba})_2(\text{dppm})_2]\text{Cl}_4$ (**9**) complex where the dppm behaves as a bridge ligand in binuclear complex. In (**1-10**) complexes the (**Btba**) ligand behaved as a bidentate through the N atom of the benzothiazole and O atom of carbonyl group.

At the minimum inhibition concentration (MIC), most of the prepared complexes appeared more effective than amikacin against *Staphylococcus epidermidis*, *E. Coli* and *Citrobacterfreundii*. The (**8**) complex, on the other side, showed activity against *Staphylococcus aureus* more than amikacin.

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