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Preparation and synthesis of a new Ligand($C_{14}H_{13}N_5O_3S$)derived from sulfamethoxazole and 2-imidazole carboxaldehyde and its complexes with Co and Ni ions , evaluating of it biological effectiveness and studying its nanoscale properties.

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Abstract

The paper presents the preparation of the Schiff base ligand of sulfamethoxazole with 2-imidazolecarboxaldehyde by the preparation of complexes with Co (II) and nickel (II) Relatedly, the samples were analyzed by UV-visible, micro-elemental, Fourier transform infrared (FTIR) approaches, ^{1}H-NMR, FESEM, X-ray diffraction (XRD), molar conductivity, magnetic susceptibility, and melting point. Additionally study the biological activities of ligands and complexes.

Keyword: Schiff base, sulfamethoxazole, UV-Visible , biological activities

1- Introduction

Schiff's base is a significant group of natural compounds. Hugo Schiff initially made them known in 1864. Schiff' bases are the buildup results of basic amines (NH_2) with C=O group mixtures. These mixtures typically contain an azomethin bunch as their major constituent, with the general equation RHC = N-R_1., where furthermore, R_1 is alkyl, aryl[1]. A number of Schiff base ligands obtained by the blend of aldehyde and an amine or ketone are valuable chelates obtained by acquiring coordination circles with blended N/O benefactors. Interest of these ligands has been driven, to a limited extent, because of potential helpful natural driven, to a limited extent, because of potential helpful natural movement of the ligands and its metal edifices counting against malarial, against bacterial, against fungal, hostile to viral action[2].

Schiff bases derived from sulfonamide drugs have attracted attention because of their structural makeup. Sulfonamide-containing compounds have been used as [3] infections Sulfamethoxazole (SMZ) is an antibiotic belonging to the sulfonamide class. Its molecular composition is $C_{10}H_{11}N_3O_3S$, and its chemical name is 4-Amino-N-(5-methyl-3-isoxazolyl)-benzene. Its molecular weight is approximately 253.279 g/mol. Domagk was the first to employ sulpha as a medication in 1932, as sulfanilamides are recognized as effective bacteriostatic agents against both gram positive and negative bacteria[4].(SMX) is somewhat soluble in ether, easily soluble in acetone, and sparingly soluble in alcohol. In a diluted sodium hydroxide solution, it dissolves.

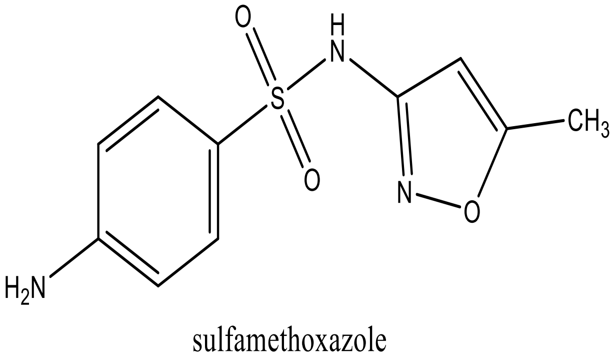


Fig. 1. structure formula of SMX

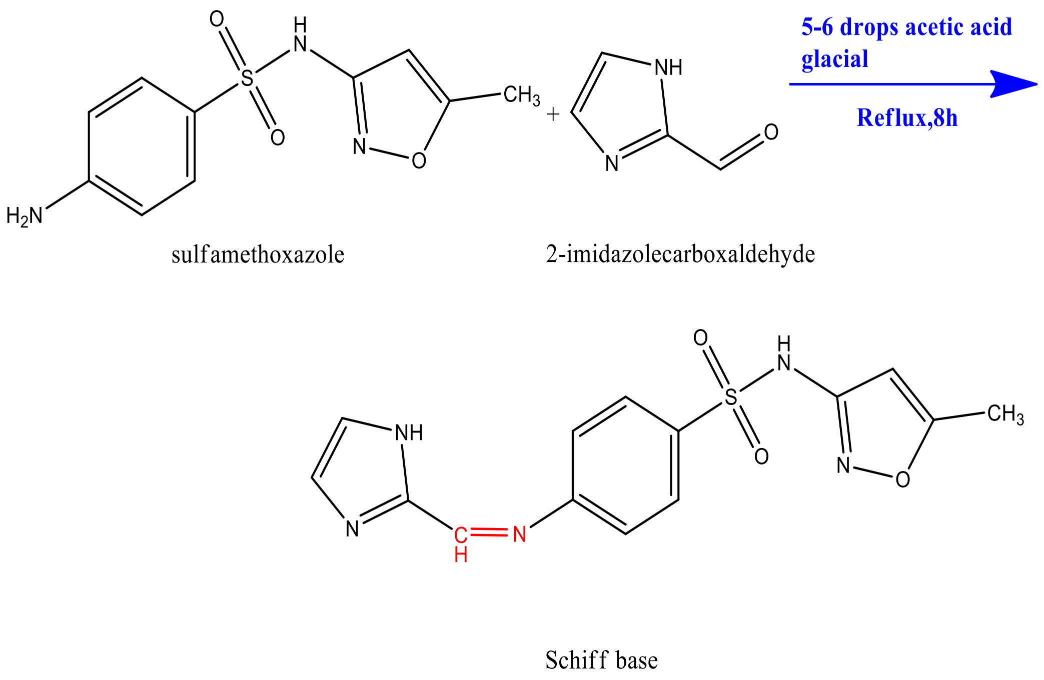
Materials and methods

Instruments, Materials and Approaches

The chemicals were sourced from Nevend New Material, Merck, and Ridel-de-Haën. Using KBr pellets and a Shimadzu spectrophotometer, FTIR spectra were captured in the 4000-400 cm^{-1} region. A quartz cell with a route length of one centimeter its used to produce electronic spectra in CH_3CH_2OH using a UV-visible spectrophotometer that covered the 200–1100 nm range. 1H NMR was acquired on a DRX (500 MHz) spectrometer in DMSO at Sharif Sainte University, Tehran, Iran. Chemical shifts were referenced to internal Me4Si. The atomic absorption spectrophotometry was performed employing a Shimadzu AA-6300 instrument to determine the metal content. Using the Stewart melting point method, the melting points of the prepared samples were determined. The magnetic susceptibilities at room temperature were measured with a Balance Magnetic Susceptibility Model (MSB-MKI). Conductivity was measured at a concentration of 10^{-3} M and at 25℃ using the Digital Conductivity Series Ino device. Cond 3110 SET1 using ethanol as a solvent. The electronic scanning measurements in the emitted field (FE-SEM) of the prepared compounds were recorded using the MIRA3 TESCAN (Czech), while the XRD measurements were made using the D_2 Phaser Bruker AXS Gmbh with an angular range of 2θ (20–80°).

Schiff base of Ligand

The ligand was made by continuously swirling the aldehyde compound 2-imidazol carboxaldehyde (0.9609 g) (10^{-2} mole) in 25 milliliters of pure ethanol. Dissolving (2.5327 g) (10^{-2} mole) of the sulfamethoxazole compound in 25 milliliters of alcohol, while stirring. The continuous solution was added , and 5–6 by dropping was added by acetic acid (glacial) to 2-imidazol carboxaldehyde. The above solutions were mixed together. The solution was raised for (8 h) at a temperature of 78 °C. The solution was cooled, and a precipitate was observed to form. Then it was filtered, dried, and recrystallized with pure ethanol (absolute). After that, the precipitate was gathered and gave a result of 95% and a m.p of(184 °C).



Scheme. (1). synthesized of the ligand

Synthesis of Complexes

The synthesis of ligand complexes was carried out by dissolving (0.331 g, 10^{-3} mol) of the ligand in 10 ml of pure ethanol, followed by the addition of (0.237 g, 10^{-3} mol) of cobalt salt dissolved in 10 ml of ethanol . Salt of nickel (0.238 gm, 0.001 mol) was dissolved in 10 ml of purity alcohol. The mixture was stirred continuously for 2 h, then cooled, resulting in making of precipitates, which were extracted from pure ethanol, dried, and then crystallized again. Precipitates of metal ion complexes that were pure and colorful were produced.

Table 1: a number of the ligand and complex's physicochemical properties

No.	Compound	Color	MP °C	Wt gm
1	**Ligand**	**Light brown**	**179-184**	**3.327**
2	$[CO(L)_2Cl\ H_2O]Cl$	**Green**	**131-136**	**1.245**
3	$[Ni(L)_2Cl\ H_2O]Cl$	**Green light**	**147-152**	**1.334**

Bioactivity

Two species of bacteria occurring in nature were used to study the biological activity of ligands and complexes: Gram-positive bacteria (Staphylococcus aureus) and Gram-negative bacteria (E. schriachia coli), using diffusion technology on the surface of culture media as antibacterial agents. The biological activity solutions were prepared at a concentration of 500 mg/L by taking the weight of 0.005 g of the ligand or the complex separately, and it was dissolved in 10 ml of dimethyl sulfur oxide

(DMSO). The compounds were incubated at 37°C for one day, and the compounds were measured the results were processed by determining the inhibition zone's diameter

Results and Discussion

Cobalt complex is a green color, and nickel is a is a light green color. but the ligand is light brown. The ligand and its complexes have the ability to dissolve in many solvents, such as CH_3CH_2OH, CH_3OH, DMSO, DMF, But the ligand and its complexes do not dissolve in pure water and do not dissolve in ether either. Here are several methods for determining the ratio of metal to ligand: the ligand-to-metal ratio [M:L]: [1:2], including the molar ratio method by June[5] and the continuous variables method presented by Job.

Molar conductivity Measurement

The molar conductivity of solutions is used in coordination chemistry to determine the properties of compounds in their solutions, and it is directly proportional to the free ions of the complex in the solution. This means the greater the number of ions released by the complex, the greater its electrical conductivity. In contrast, a complex that does not ionize will have a low conductivity, meaning that the complex is non-ionic in solution[6,7]. The molar conductivity of the prepared chemical compounds was determined to clarify the formula of the coordination compounds and distinguish the ions as to whether they are located inside the coordination sphere or outside it[8]. The solvent DMSO was used at a 10^{-3} M concentration at 25℃ temperature, and the molar conductivity measurements match the proposed formula.. It was found that the cobalt and nickel complexes have a molar conductivity of the same magnitude (42.1 ,35.8 $ohm^{-1}.cm^{2}.mole^{-1}$) , respectively. According to these findings, the complexes exhibit an ionic nature with a [1:1] ratio.

Table . 2. molar conductivity $\Omega^{-1}.cm^{2}.mole^{-1}$ from complex of Ligand

No.	Complex	(Ω^{-1},cm^2, $mole^{-1}$)	Ionic Ratio
1	$[CO(L)_2Cl\,H_2O]Cl$	42.1	1:1
2	$[Ni(L)_2Cl\,H_2O]Cl$	35.8	1:1

The ^1H_NMR Specttrum of the Ligand

The spectra of proton (1H-NMR) for the synthesized ligand (C14H13N5O3S) were analyzed. The spectrum displayed several distinct signals. A single signal at δ = 4.5 ppm with a spin count (S) of 3H was attributed to the methyl group (CH_3) linked to the oxazole ring. Another single signal at δ = 7.3 ppm (S, 1H) was identified as originating from the CH-group of the azomethine moiety. Multiple signals ranging from δ = 7.5 to 7.9 ppm (M, 7H) were observed, corresponding to the protons of the aromatic ring. Additionally, a singular signal of δ = 8.4 ppm (S, 1H) was attributed to proton of the secondary amine in the imidazole ring. Furthermore, a peak at δ = 6 ppm (S, 1H) was indicative of the proton of the secondary NH group attached to the oxazole ring. Notably, signals at a chemical shift of δ = 2.51 ppm were identified as arising from the protons (H) by DMSO-d^6 solvent [9].

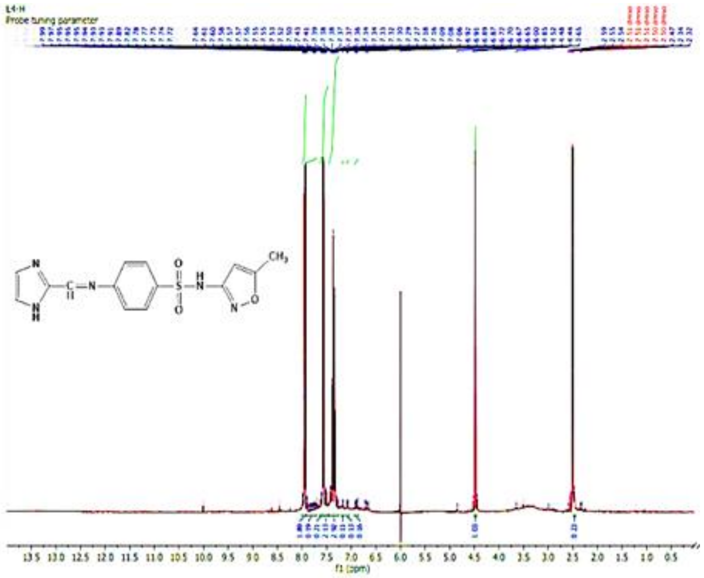


Fig. (2). ^{1}H-NMR for the Ligand($C_{14}H_{13}N_5O_3S$)

Table (3) Spectrum signal values (1H -NMR) for the Ligand($C_{14}H_{13}N_5O_3S$)

^{1}H-NMR	
Signals (ppm)	**Assignment**
S, 3H , δ =4.5	**-CH_3**
S, 1H , δ =7.3	**-CH (azomethine group)**
S, 1H , δ =8.4	**-NH(2^oamine) Imidazole ring**
M, 7H , δ =7.5-7.9	**C-H (aromatic ring)**
S, 1H , δ =6	**-NH (2^oamine)**
δ =2.51	**DMSO-d^6**

Infrared spectra of ligand and their complex cobalt (II) and nickel (II) complexes

A Shimadzu 8400S device was used to measure the infrared spectrum using potassium bromide (KBr) to determine the active groups of the ligand. The spectrum of the ligand showed a number of bands, the most important of which is the secondary amine group (NH) υ at the wavenumber 3240.19 cm^{-1}. A new band showed in the ligand spectrum, the appearance of which shows that the azomethine ligand, which is a part of the Schiff base, is forming, which appeared at the wave number 1620 cm-1, and the carbonyl group disappeared during the reaction. The appearance of other bands at 3070 cm^{-1} and 2977–2877 cm^{-1}) belonging to (-CH) υ aryl ring and aliphatic group, respectively[10]. For the (C=C) υ aryl groups, It has bands at 1535-1465. cm^{-1}. The (C=N) υ group of the oxazole ring appeared as a band in the ligand spectrum at 1596 cm^{-1}. The spectra of the synthesized complexes were observed with compared the spectra of the only ligand. It was determined displacement of some bands and the appearance of new bands, all of which are considered evidence of the coordination process between the metal ion and the ligand occurring, which can be explained as follows: The absorption band of the secondary amine group (N-H) υ is shifted towards smaller frequencies in the spectrum of the complexes when compared to the spectrum of a specific ligand, as it is shifted by an amount of 39–

100 cm^{-1} . New bands appear at frequencies (570 cm^{-1}). These bands return to M-N υ groups as a result of the N atoms in the secondary amine and azomethine groups coordinating the ligand with the metal ions. The appearance frequencies are 3348–3355 cm^{-1}. These frequencies are due to the OH groups in the water of crystallization molecules in all complexes[11]. The C=N group of the Schiff base, which was visible at (1620) cm^{-1} in the free ligand's spectra, was moved 30 cm^{-1} following coordination with the complex spectra, indicating a shift towards lower or higher frequencies. with the metal ions[12,13].

Table 4: Important spectral bands of Ligand, Co (II),and Ni (II) Complex in IR

compounds	**υ(O-H) Water molecules**	**υ(N-H) 2^oamine**	**υ(C-H) aromatic**	**υ(C=N) ring**	**υ(C=N) Imine**	**υ(C=C) aromatic**	**υ(M–N)**
Ligand	-	3240	3070	1596	1620	1535 1465	-
[CO(L)$_2$Cl H$_2$O]Cl	3348	3224	3093	1620	1650	1500 1473	570
[Ni(L)$_2$Cl H$_2$O]Cl	3456	3139	3093	1620	1650	1500 1496	570

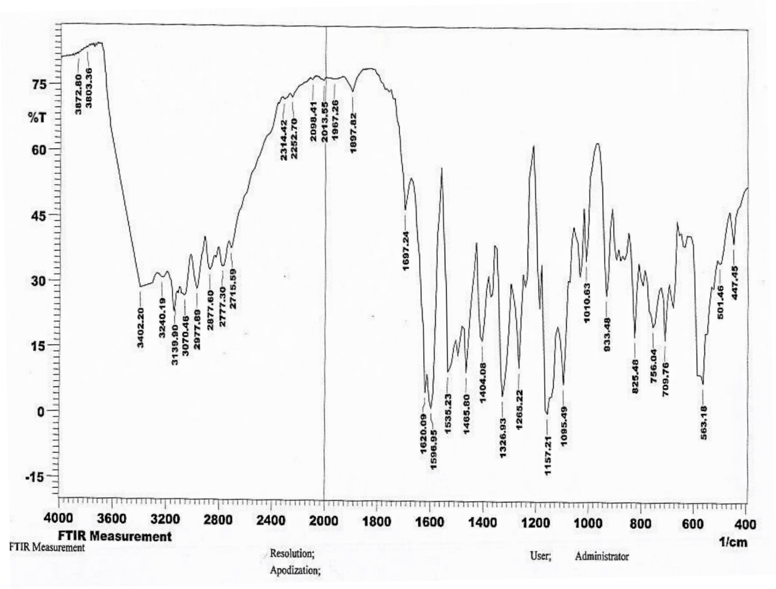


Fig. (3). FT-IR of Ligand

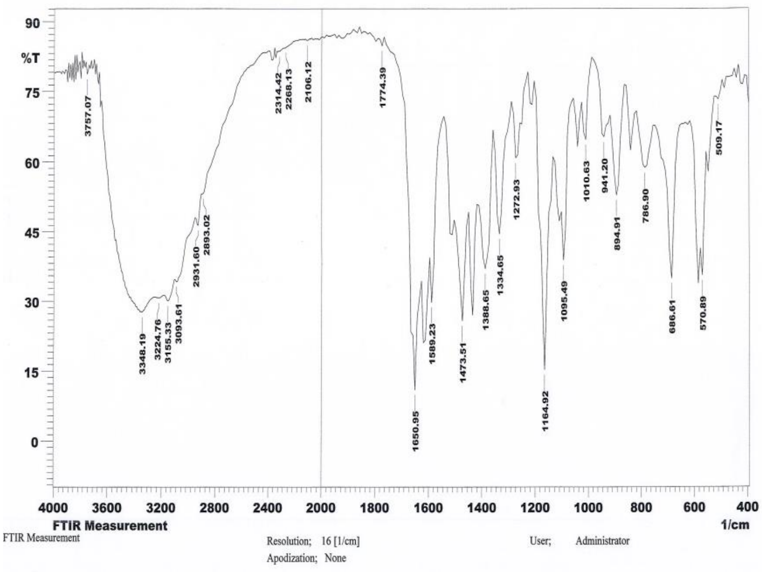


Fig. (4). FT- IR of cobalt (II) complex

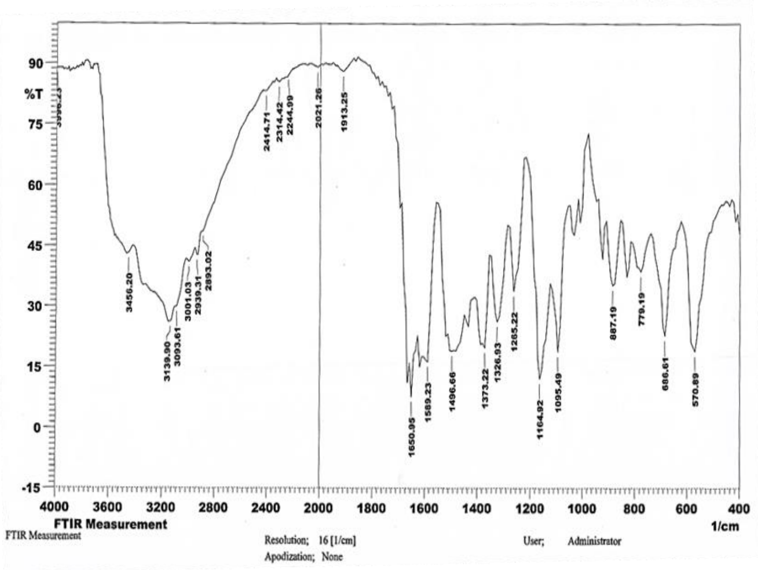


Fig. (5). FT- IR of Nickel complex

Electronic Spectra

The electronic spectra of the synthesized Ligand($C_{14}H_{13}N_5O_3S$) nanocomposite complexes were analyzed as follows:

Looking at the electronic spectrum of the ligand in Figure (6), the ligand spectrum showed 3 peaks in 208 nm(49019 cm^{-1}), 208 nm(41322 cm^{-1}), and 336 nm(29761cm^{-1}). The azomethine group, C=N, has transitions (π-π*) represented by the first and second peaks, and transitions (n-π *) by the third peak ,Cobalt complex appears in Fig (6) with five absorption peaks in its spectrum. The 3 peaks at 227 nm(44052 cm^{-1}),266 nm (37593 cm^{-1}),452 nm (22123 cm^{-1}) be a part of the ligand field, whereas the 580 nm peak refers to the transition $^1A_1g \rightarrow {}^1T_2g$,, and the peak at 650 nm refers to the transition $^1A_1g \rightarrow {}^1T_1g$ [14,15]. As for the nickel complex in Fig. 6, it showed absorption peaks at 220 nm cm^{-1}, 236 nm (42372 cm^{-1}), and 335 nm (29850 cm^{-1}). All of these peaks indicate the ligand field. As for the peaks at 450 nm (22222 cm^{-1}), 660 nm (15151 cm^{-1}), and 802 nm (12468 cm^{-1}), which indicate transitions $^{3}A_{2}g(F)\rightarrow{}^{3}T_{1}g(F)$, $^{3}A_{2}g(F)\rightarrow{}^{3}T_{1}g(p)$, $^{3}A_{2}g(F)\rightarrow{}^{3}T_{2}g(F)$, it showed that the shape is regular octahedral[16].

Table. (5). peaks absorption values, magnetic momentum and expected geometry for ligand and their complexes Co(II), Ni (II)

Com	λ (nm)	υ^{-} (cm^{-1})	Transitions state	Geometry	μeff (B.M)
Ligand	208 242 336	49019 41322 29761	(π-π*) (π-π*) (n-π *)	-	-
[CO(L)$_2$Cl H$_2$O]Cl	227 266 452 580 650	44052 37593 22123 17241 15384	Intra Ligand Intra Ligand Intra Ligand $^{1}A_{1}g\rightarrow{}^{1}T_{2}g$ $^{1}A_{1}g\rightarrow{}^{1}T_{1}g$	Octahedral sp^{3}d^{2} distorted	3.76 (Para.)
[Ni(L)$_2$Cl H$_2$O]Cl	220 236 335 660 450 805	45454 42372 29850 15151 22222 12468	Intra Ligand Intra Ligand Intra Ligand $^{3}A_{2}g(F)\rightarrow{}^{3}T_{1}g(F)$ $^{3}A_{2}g(F)\rightarrow{}^{3}T_{1}g(p)$ $^{3}A_{2}g(F)\rightarrow{}^{3}T_{2}g(F)$	Octahedral sp^{3}d^{2} Regular	2.75 (Para.)

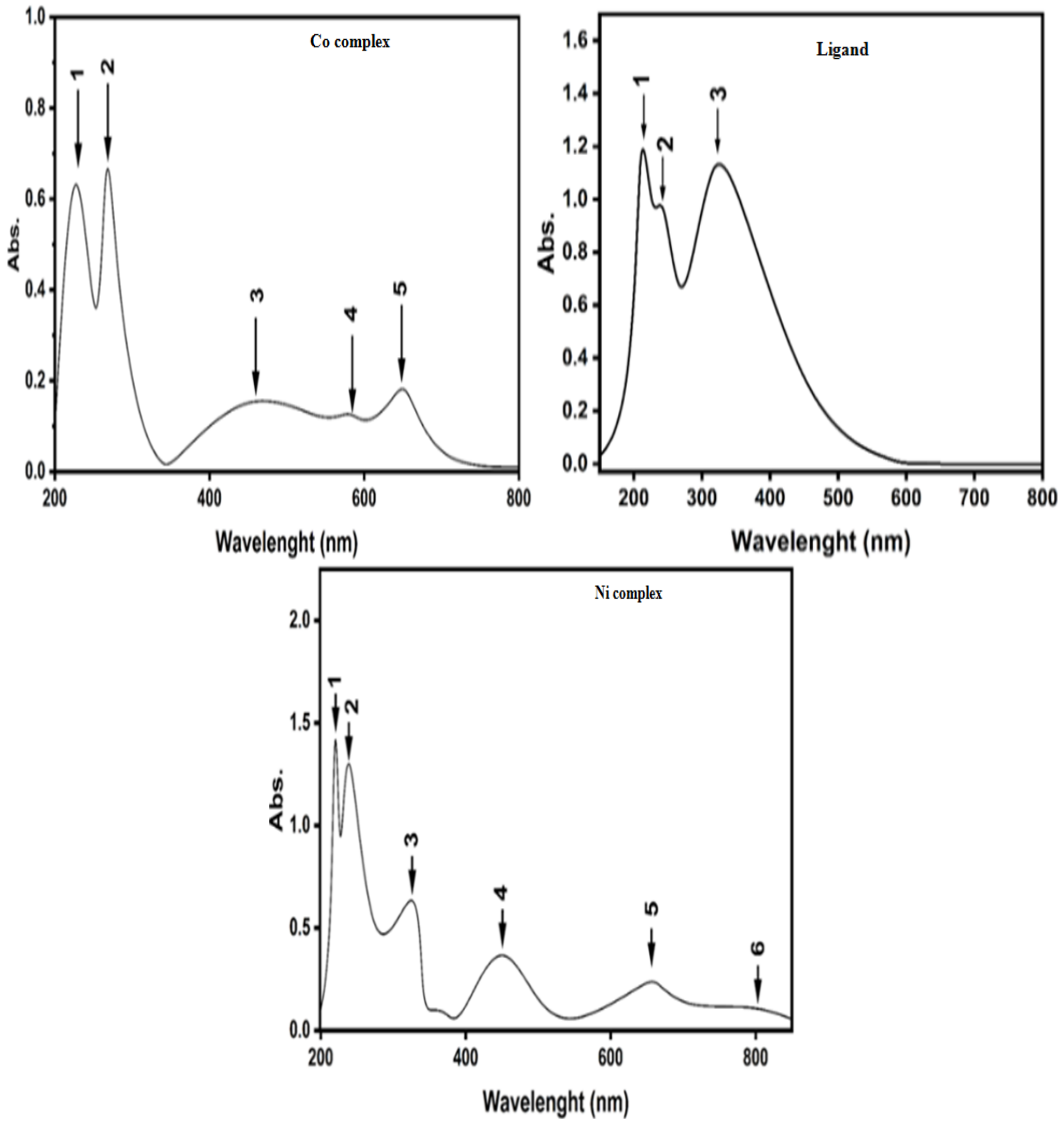


Fig. (6). Uv-Vis. Spectrum of ligand and complexes of Co(II) and Ni(II)

magnetic susceptibility

The magnetic moment values of the synthesized metal complexes were measured at 25 °C. The magnetic moment of the complex of cobalt (II) is equal (3.76 B.M), which is a value that is compatible with the presence of three individual electrons and has an electronic arrangement of $t_{2}g^{5}eg^{2}$ and $sp^{3}d^{2}$ hybridization. Its geometric shape is a distorted octahedral with a high spin[17]. The nickel complex showed a magnetic moment equal (2.75 B.M) as a result of the presence of two electrons and the $sp^{3}d^{2}$ hybridization, while its electronic arrangement is $t_{2}g^{6}$ eg^{2}[18]. .The results are given in Table 5.

FESEM Microscope

Field-Emission Scanning Electron Microscope to measure and analyze the surface and properties of the ligand and their metal prepared from cobalt (II) and nickel (II), which depend mainly on the nature of the surface, the cross-section distance of 200 nm, while the magnification power of Mag = 135 KX.

Through FE-SEM analysis, the ligand was found to be in the form of spherical granules with a mean particle size of 31.56 nm,, while FE-SEM analysis of the cobalt (II) complex showed it to have spherical granules with agglomerates and a mean particle size of 43.73 nm.. The FE-SEM of nickel (II) also showed that it is complex; it is in spherical granules with 43.93 nm particle size. The above result showed that the prepared ligand and the cobalt (II) and nickel (II) complexes are characterized by particle sizes less than 100 nm[19], i.e., within the nanoscale. These results enabled us to study the compound ligands Co (II) and Ni (II) in the field of medicine, their ability to eliminate many types of pathogenic bacteria, including E. coli and Staphylococcus aureus, and the probability of using them as medicine, and this should be clarified later in this study[20,21].

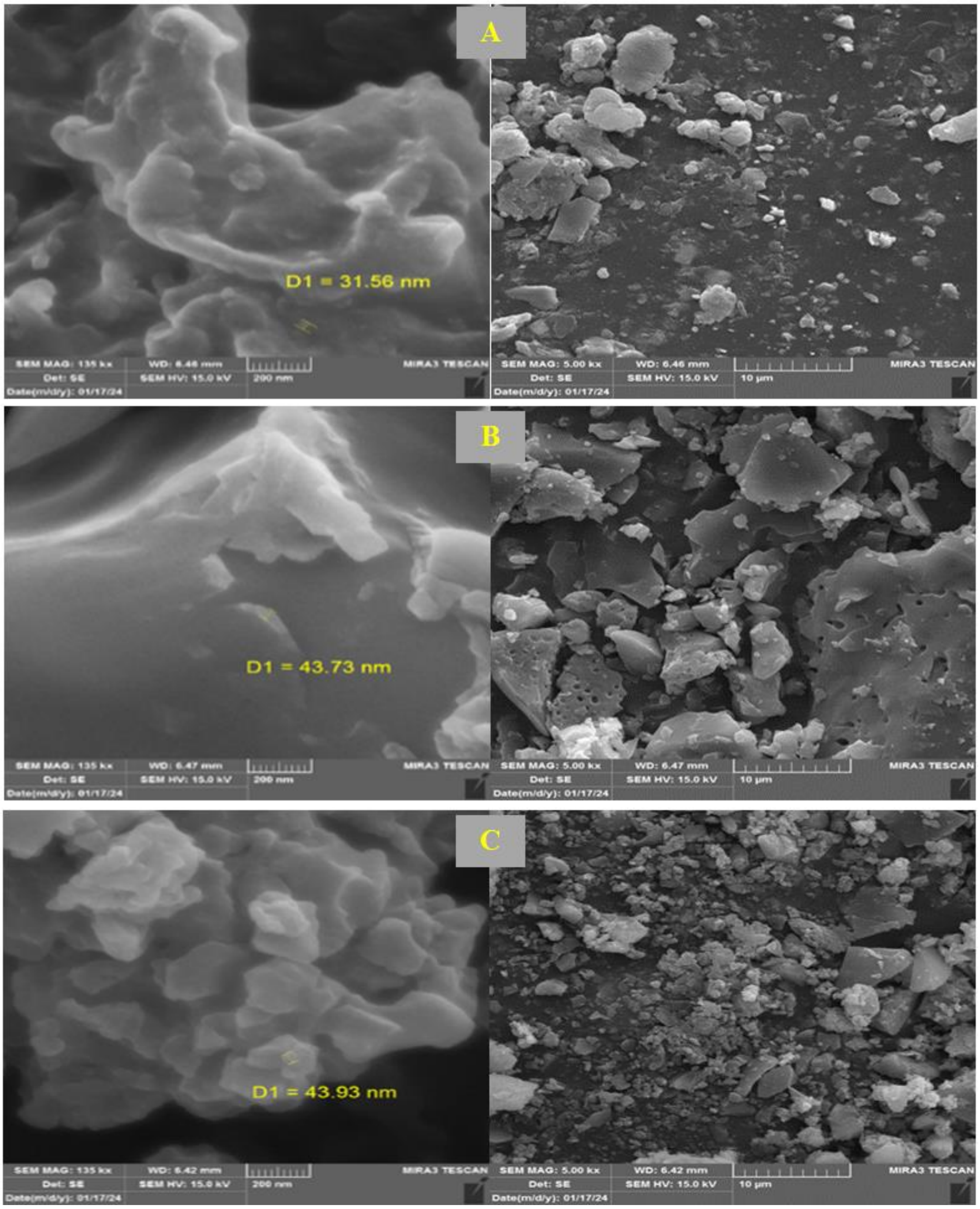


Fig. (7). FE-SEM images of the prepared A) ligand, B) Co Complex and C) Ni complex

X-ray diffraction (XRD)

Using X-ray Pattern within the (the range of angular is 10°–80°), the ligand and its metal complexes, cobalt (II) and nickel (II), in their solid state were studied to determine some structural characteristics, crystal sizes, and crystal structures. Additionally, the density of dislocations and microstresses was computed. density) when transforming the ligand into metal complexes To be able to assess the degree of purity and the crystal structural flaws. A review takes place for a number of diffraction peaks for various reasons, such as the macrostrain, which includes the absence of crystal lattice deformation and crystal cracking (faulting) brought on by crystal distortions, the crystal's domain size, and the distribution of domains[22]. The crystal sizes of the ligand and their metal complexes, nickel and cobalt (II), were also determined using the Debye-Scherrer equation, as follows.

$$D = \frac{K \ \lambda}{\beta \cos \theta}$$

D equal average crystal size.

k equal the shape factor (0.9).

λ represents length of wave X-rays, with CuKα = 1.54056 A°.

β equal total width half maximum height FWHM

ϴ equal is the angle of deviation

Through X-ray measurements by Fig 8, the ligand, cobalt (II), and nickel (II) complexes are of various structures, and this indicates the occurrence of coordination between metal ions and the ligand and formation of complexes. X-ray diffraction measurement showed that all the prepared compounds have a crystalline structure because all the peaks are sharp and all the resulting materials are of a nanoscopic nature. From the results obtained by X-ray diffraction, it was found that all the prepared materials are nanoscale in nature, as the size of the crystals is less than 100 nanometers. These findings support our observations of earlier (FESEM) examinations and are inside the nanoscale..

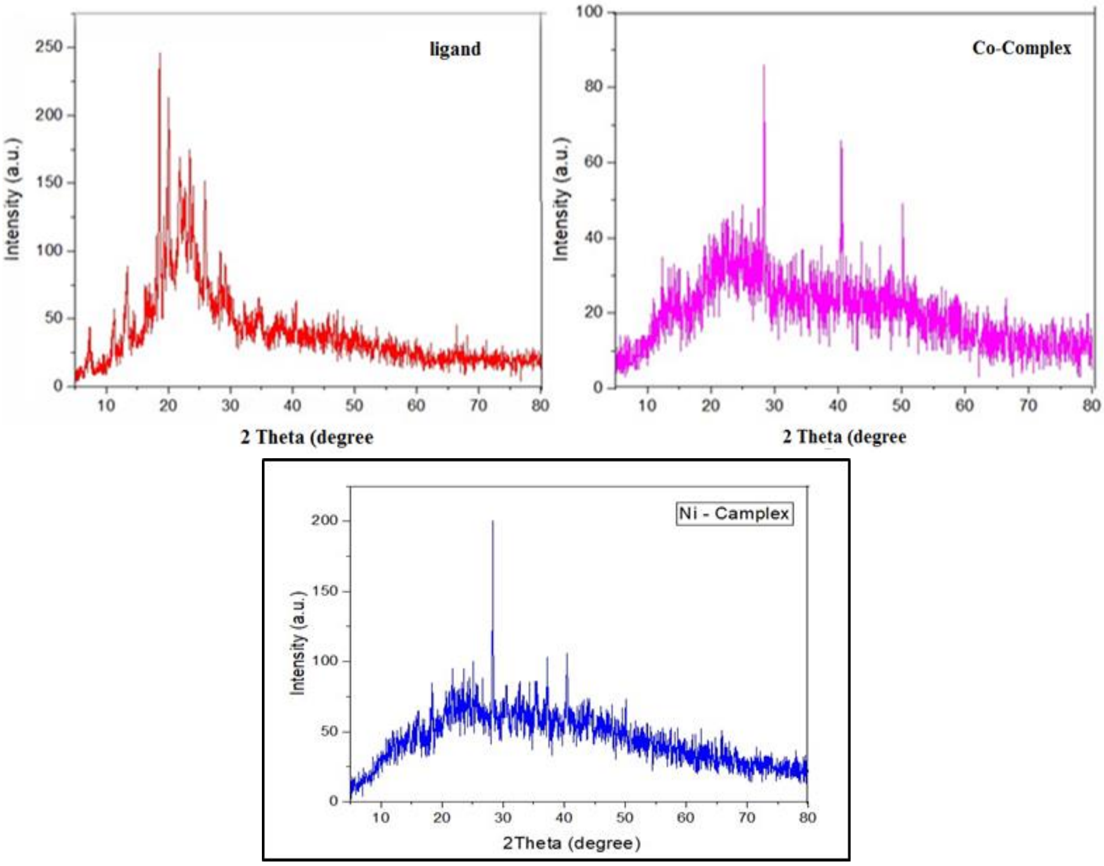


Fig. (8). XRD patterns of ligand and its prepared metal copmlexes

Table(6) Interplanar distances and the 2 theta of peaks, intensity for ligand and complex

Compound	No.	Pos. °2Θ.(Radian)	d-spacing A°	Peak Width (FWHM)	D Crystallite size(nm)	Intensity In	Rel. Int [%]
Ligand	1-	18.775	4.985	0.196	42.92	278	100%
	2-	20.339	8.548	0.157	53.71	259	93%
	3-	21.267	6.202	0.472	17.89	290	58%
	4-	23.423	4.497	0.236	35.92	288	96%
	5-	25.897	3.440	0.236	36.09	288	51%
Co complex	1-	28.374	3.142	0.157	54.53	99	100%
	2-	40.867	2.225	0.236	37.54	100	69%
	3-	50.230	1.814	0.90	10.19	75	91%

Ni complex	1-	28.424	3.150	0.231	37.07	265	100%
	2-	37.381	2.407	0.275	31.87	104	75%
	3-	40.421	2.053	0.221	40.03	90	25%

Pharmacology Results

Antibacterial activity

In this research, the biological activity of ligands and complexes prepared from their solution dissolved in DMSO was evaluated against two dangerous bacterial species: the gram-positive Staphylococcus aureus and the gram-negative Escherichia coli. The result obtained showed that the cobalt (II), at the same concentration (500 ppm), had relatively high activity compared to the ligand against Escherichia coli bacteria, while the nickel (II) complex gave the same activity compared to the ligand against the bacteria under study. The activity of compound nickel (II) at a concentration of 500 ppm was highly effective against bacteria (Staphylococcus aureus), while the percentage of ligand and cobalt (II) complex at the same concentration showed moderate activity compared to the nickel (II) complex against the same bacteria under study[23].

Table (7) show the inhibition range of each of ligand and its complexes against some bacteria

NO	**Complex**	**Corresponding**	
		E. coli	**S. aureus**
		ppm 500	**ppm 500**
1	**Ligand**	**5**	**5**
2	**$[CO(L)_2Cl\ H_2O]Cl$**	**7**	**5**
3	**$[Ni(L)_2Cl\ H_2O]Cl$**	**5**	**6**

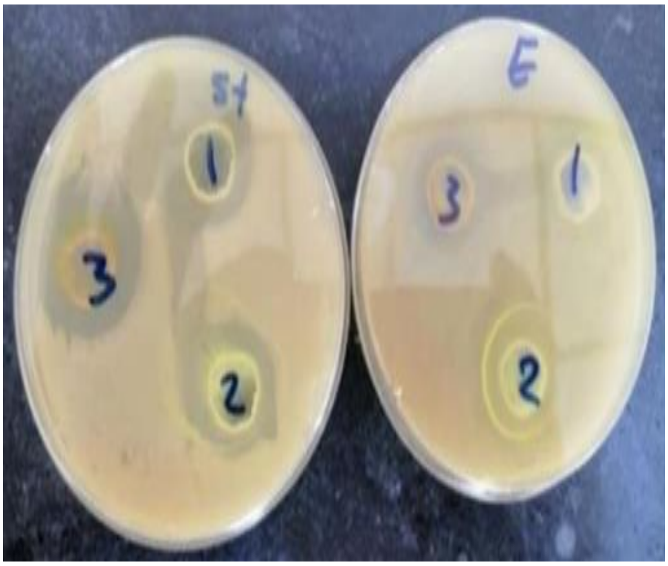


Fig. (9). Iinhibiting of ligand (1), cobalt(2), nickel (3) complexes, on types S. aureus and E. coli (500 ppm)

Conclusions

In this research, the ligand was prepared from sulfamethoxazole with 2-imidazolecarboxaldehyde, and Co (II) and Ni (II) complexes of this ligand were prepared by reacting with divalent aqueous cobalt chloride and divalent aqueous nickel chloride. Ligands and their complexes were characterized in a variety of spectroscopic analyses. By the physical method, infrared spectra confirmed the formation of the azomethine group. These spectra also showed that the compound is coordinated with cobalt and nickel metals through the nitrogen atom of the azomethine of the Schiff base and the nitrogen imidazole ring. Upon coordination, the azomethine group shifted towards higher frequencies than in the free compound. indicating the creation of metal complexes. The electronic spectrum of the cobalt (II) complex and nickel (II) complex indicates an octahedral geometric shape. Molar conductivity measurements showed that cobalt (II) and nickel (II) have an ionic nature. XRD measurements and FE-SEM analysis demonstrated the ligand and its complexes are nanosized. By testing the two types of bacteria, negative Ekram and positive Ekram for each of ligand and cobalt (II) and nickel (II) showed promising antibacterial activity.

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