Synthesis, Spectral and Biological studies of Some transition metals with New Heterocyclic Imidazole Azo Ligand

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ABSTRACT

The new azo reagent has been synthesized by reaction of imidazole and 4,4'-sulfonyl dianiline . This reagent has been characterized by different techniques such as, (IR, U.V-Vis) spectroscopy, (^{1}H, ^{13}C) NMR spectra are used to produce of the azo reagent. Its (Co^{2+}, Ni^{2+} and Cu^{2+}) binuclear complexes were prepared and characterized by (IR, U.V-Vis) spectroscopy, atomic absorption, conductivity measurements and magnetic quantifications. Nature of the complexes produced have been studied followed the mole ratio study and continuous contrast methods, Beer's law followed during the concentration scope (1×10^{-6}-1×10^{-4})M. The reagent acts as binuclear and bidentate at the same time with (N,N) set donar and the coordinates through nitrogen atom from azo group and nitrogen atom(N3) from imidazole ring . Octahedral geometry structure for Co(II), Ni(II)and Cu(II) complexes were suggested. All these complexes and the reagent were evaluated against two kinds of human pathogenic bacteria such as Staphylococcus aureus (Gram Positive) and Escherichia coli (Gram negative). 7733

KEYWORD: Azo compound, Complexes, Imidazole, Ligand, Dapsone.
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INTRODUCTION

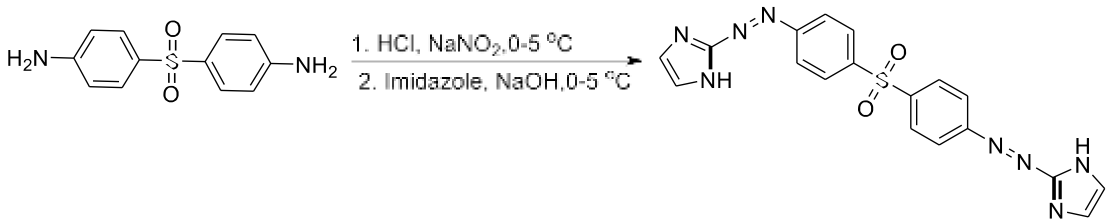
Imidazole is an important type of the aromatic compounds heterogeneous ring, as it enters in many areas, including chemical and the biological, due to the high therapeutic property of an imidazole and the wide range of its clinical drugs As the researchers proceeded to prepare a large number of chemical treatments, including Antioxidant, Antifungal [1], antimicrobial[2], anti-inflammatory[3] anticancer[4] and anti-tumor agents[5]. An imidazole ring is present in many natural products and in the human body, among which are the vitamin B7 or biotin[6], and there is an imidazole ring in medicines to treat certain diseases such as cimetidin [7]which is used. In the treatment of stomach and duodenal wounds and the drug Metronidazole, or as it is known commercially as Flagyl[8, 9]. In this study, we report the preparation, identification of new ligand (SBPDBI) and its complexes Co (III), Ni (II) and Cu (II) ions. The synthesis compounds were studied by various spectral analysis and screened for their biological activities against Klebsilla pneumonia, streptococcus as antibacterial and Aspergillus Niger. as antifungal. Moreover, the study of Cu (II) complex of prescription drug anticancer by using the lines of breast cancer cells and compared with line of the normal cells by MTT assay.

Experimental Part Apparatus and materials

All chemicals and solvents were highest purity obtained from Fluka, The Merck and BDH. The melting points were determined on an Electro thermal, melting point 9300. IR- spectrum have been taken at a Shimadzu, FTIR- 8400S Fourier Transform Infrared Spectrophotometer at the (4000- 400) cm^{-1} spectrum areas for models produced like KBr discs. Molar conductance measurements were determined in DMSO (10^{-3} M) at room temperature HANNA model 214EC conductivity meter the pH measurements were carried out using pH-meter Hanna. The metal contents of the complex was measured by using atomic absorption technique by Perkin-Elmer model 2280.

Preparation of the ligand

The new azo ligand was created in relation to the Gusev method [10] by dissolving (2.4830 g g, 0.01 mol) of 4,4′-sulfonyl dianiline in a mixture consisting 25 mL of distilled water and 3 mL of concentrated hydrochloric acid. A solution has diazotized at (0–5) ^{0}C 10 mL of aqueous (0.69 g, 0.01 mol) sodium nitrite. Consequentially, diazonium chloride solution was added to the mixture drop by drop with stir then, Imidazole (0.68 g, 0.01 mol) was dissolved in 150 mL of alkaline ethanol and then cooled lower than 0 0C. Next to effecting addition, then left the content for two hours, then 150 mL of cooled distilled water was add, the pH of the solution maintained at 7 by HCl, brown molecules were precipitated and left for 24 h. The precipitate has been filtered off and washed up many times with (1:1) (ethanol : water) mix then recrystallized two times from hot ethanol, and dried in a vacuum desicator.



Scheme1: Preparation reaction of the azo ligand (SBPDBI)

Preparing the Buffer Solution

(0.01mol/L, 0.771 gm) for ammonium acetate was melted at one liter for doubly deionized water. For only pH scope (4-9) was the use of CH_3COOH or NH_3 solution.

Preparation the Standard Solution

The metal salt solutions ($1x10^{-3}$ -$1x10^{-6}$ M) have organized through dissolving the fitting weights of every of these salts [$CoCl_2.6H_2O$, $NiCl_2.6H_2O$ and $CuCl_2.2H_2O$] in the equipped buffer solutions. Simultaneously, the range of concentration has employed for preparing the ligand solutions obtained via dissolving (0.04 g, 0.001 mole) from the ligand (SBPDBI) in (100) mL of ethanol. The solution was diluted to obtain concentrations ($1x10^{-3}$ -$1x10^{-6}$ M).

Determination of Optimum Concentrations

The optimum concentrations for the mixing process, the solutions ranged between ($1x10^{-3}$ - $1x10^{-6}$ M) were determined for both the ligand and metal ions. Equal volumes were mixed for both the ligand and metal ions at rang of acidity functions (3-13) and measured the absorption value for these solutions. The high-ranged concentrations ($1x10^{-3}$ and $6x10^{-4}$ M) showed complex precipitation for instant mixing of these solutions. So the concentrations between ($1x10^{-4}$ - $5x10^{-4}$ M) were chosen because it gave acceptable absorption and some of them comply with the Beer - Lambert law, as for the minimum concentrations ($1x10^{-5}$ and $1x10^{-6}$ M), they gave a weak measured.

Determination of Optimum Acid Function

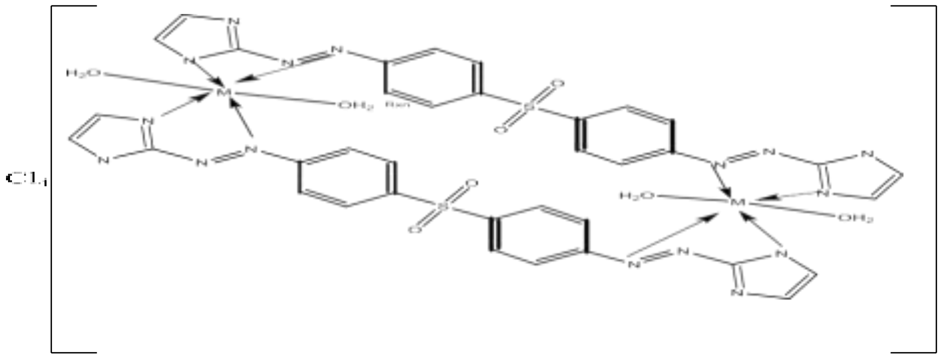
The effected of acidic function change was studied at a range of pH (3-12) to reach the optimum acidic function of the metal ions after the optimal conditions were established, which included the determination of the optimal concentrations of ligand and metal ions.

Determination of Standard Calibration Curve of the Metal Complexes

A solutions ranged between ($1x10^{-4}$ -$5x10^{-4}$M) for both the ligand and the metal ions studied, the equal volumes of same concentration of metal salts solutions and ligand solutions were mixed at optimal acidic function and the wavelength that gives the highest absorbance (λmax). The absorption values of these solutions in practical experiments showed that the optimal concentrations for complexes prepared and suitable for spectral measurements fall within the range ($1x10^{-4}$ - $5x10^{-4}$ M) were obeyed the Lambert-Beers law. According to the graphs plotted of the standard calibration curve for the metal ions complexes solutions, the relationship between the ligand and metal ions concentrations was obeyed the Lambert-Beers law.

Synthesis of Complexes

The complexes have equipped through adding dropwise based on hot stirring ethanolic solution of (2 mmole) ligand to stoichiometry amount of M:L ratio of (2:2) of [Co(II), Ni(II) as well as Cu(II)] that dissolved in a primed buffer solution at finest pH. A mix has heated up under (60°C) and stirred up to (1 hour). After that, at room temperature, they have left to cool. A colored precipitate has filtered, washed many times with a (1:1) (water: alcohol) mix then left to desiccated in vacuum desiccator. A recommended stoichiometry structure for the complexes has been illustrated in scheme (2).



Scheme 2: The proposed molecular structure of complexes, M= Co(II), Ni(II) and Cu(II)

Antimicrobial Activity Study

Antibacterial activities of the new Azo ligand and its metal complexes were screened by Kirby-Bauer disk diffusion method [13]. This paper employs dual strains for pathogenic bacteria first, a Staphylococcus aureus (Gram Positive) and the other strain, Escherichia coli. (Gram Negative). The used biochemical solutions were organized through dimethyl sulfoxide (DMSO) as solvent, where a single concentration (C) $1x10^{-3}$M is joined. The dishes have been incubated under 37°C for a complete day. Based on the inhibition zones, the diameter (Inhibition area) mm has formed after a complete day as a principle for an intensity of the influence of synthetic chemical compounds on a growth of specific cultivated bacteria strains.

Consequences and Discussion General

The Azo ligand (SBPDBI) is Orange, which is not soluble in water and soluble in common organic solvents. The reaction of this ligand with the metal ions gives different color crystals. All complexes are quiet water - insoluble, stable in air, while they are soluble in most organic solvents such as DMSO, DMF, acetone…etc. Physical Properties and Elemental Analyses Reacting the organized ligand (SBPDBI) with a selection of metal ions [Co(II), Ni(II) and Cu(II)] at finest concentration and pH, that causes formed complexes with formula [[Co_2 $L_2.4H_2O$] Cl_4, [Ni_2 $L_2.4H_2O$] Cl_4, [Cu_2 $L_2.4H_2O$] Cl_4]. Elemental analysis is in agreement with formula of the ligand (SBPDBI) and its complexes given in (Table 1).

Table 1: Physical details and analytical information of the made azo ligand (SBPDBI) besides its complexes.

Empirical formula (M . Wt) gm/mol	pH	Mp., °C	Color
($C_{17}H_{14}N_4O_5S$)	----	193-195	Orange
[$Co_2L_2.4H_2O$]Cl_4	10	234-237	blackish-brown
[$Ni_2L_2.4H_2O$]Cl_4	8	218-223	reddish-brown
[$Cu_2L_2.4H_2O$]Cl_4	8	224-226	dark red

Influence of pH

The pH magnitudes intended for the metallic ions complexes solutions have been investigated for detecting the finest pH medium for preparing the metallic ions complexes, the best concentration of complexes have reserved as provided an ascension to a study (λ_{max}) at numerous pH magnitudes. The pH influence has been as well examined within (3-12) range. Figure (1,2,3) depicted an absorbance for the pH curves that had completed at (λ_{max}) and some the concentrations under study for every metal ion solution. The huge band of the curves for pH which was reflected as a reference of complex formation and acceptance as finest pH for preparing complexes. Hence, we deduce that all equipped chelate complexes with a selection of metal ions have been prepared in basic medium[14,15].

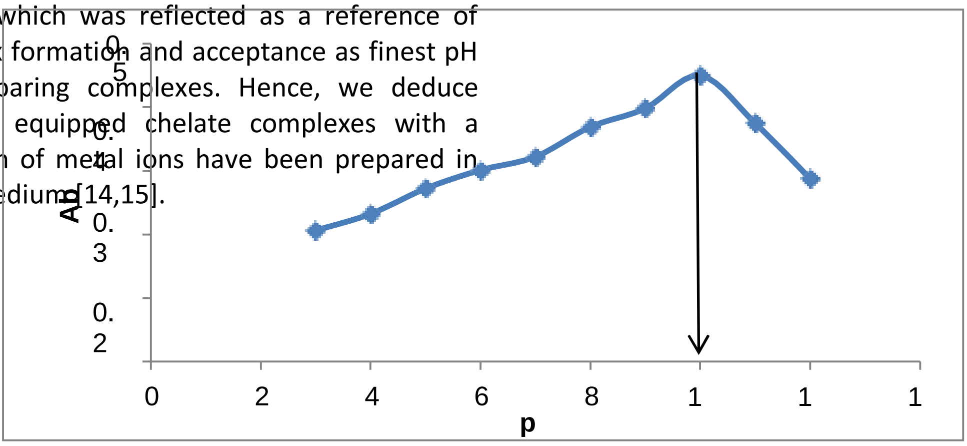


Fig. 1: Influence of the acidic function (pH) on the absorbance for [Co2 L2.4H2O] Cl4 at concentration (1 x 10^{-4} M) and λmax(440 nm)

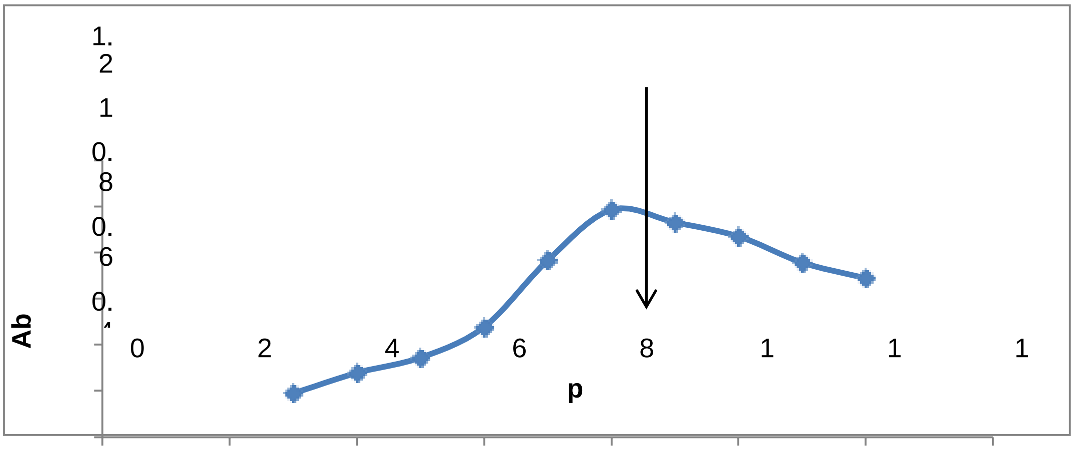


Fig. 2: Influence of the acidic function (pH) on the absorbance for [Ni2 L2.4H2O] Cl4 at concentration (1 x 10^{-4} M) and λmax(445 nm)

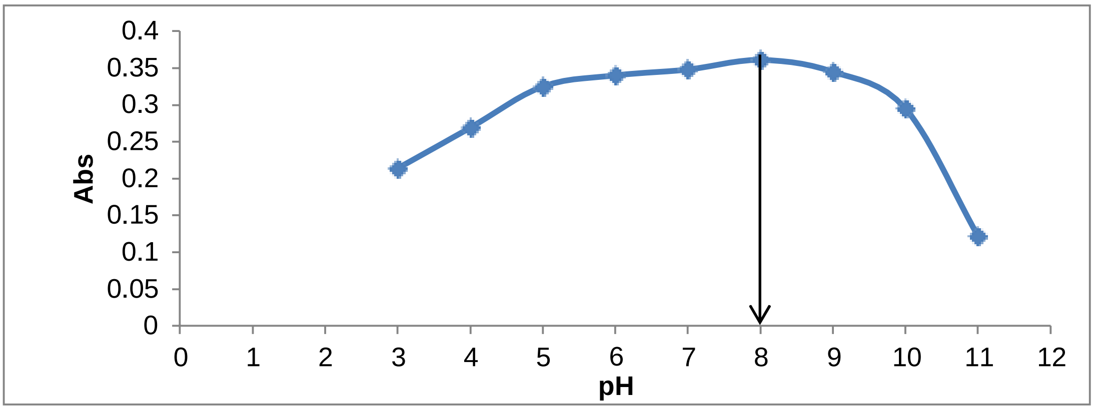


Fig. 3: Influence of the acidic function (pH) on the absorbance for [Cu_2 L_2.$4H_2O$] Cl_4 at concentration (1×10^{-4} M) and λ_{max}(450 nm)

Mole Ratio [Metal:Ligand] Ratio

The mole ratio technique was adopted with the intention of identifying spectrophotometrically of a composition of metal chelate complexes and for conclusion of potential structural formula of organized metallic ions complexes, this technique has been in accordance with an range measurement for every complexes solution under fixed finest pH and concentration at a supreme absorbance wavelength. The colors intensity for the solution of equipped metallic ions complexes has raised based on intersection point for [M:L] ratio approach. The continual stability of color beyond an intersection point signifying the formed metallic complexes [15]. The structure of formed complexes in solutions was proven by mole ratio method. Accordingly, the consequences show a mole ratio [M:L] [2:2] for entire metal ions to ligand ratio. The method supports the complexes formation, as explained by Figure (4).

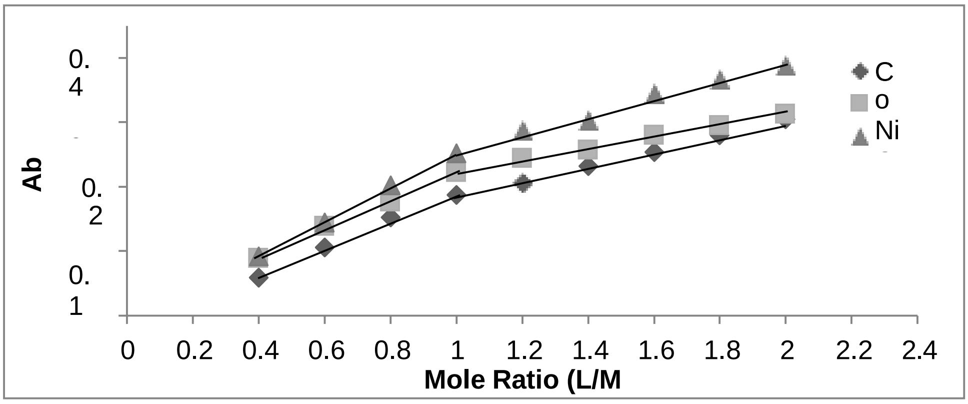


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Fig. 4: The mole ratio plot of metal ions(SBPDBI) ligand complexes solution at optimum pH and λmax.

Calibration curve

Varied molar concentration (1×10^{-3}-1×10^{-6}M/L) for mixed aqueous-ethanol ligand and metal ions, only reach (1-8$\times10^{-5}$ M/L) concentration followed Beer's law as well showed obvious intensive color. The best straight lines fit have been taken for correlation factor R>0.9980 like assigned at Fig. 5

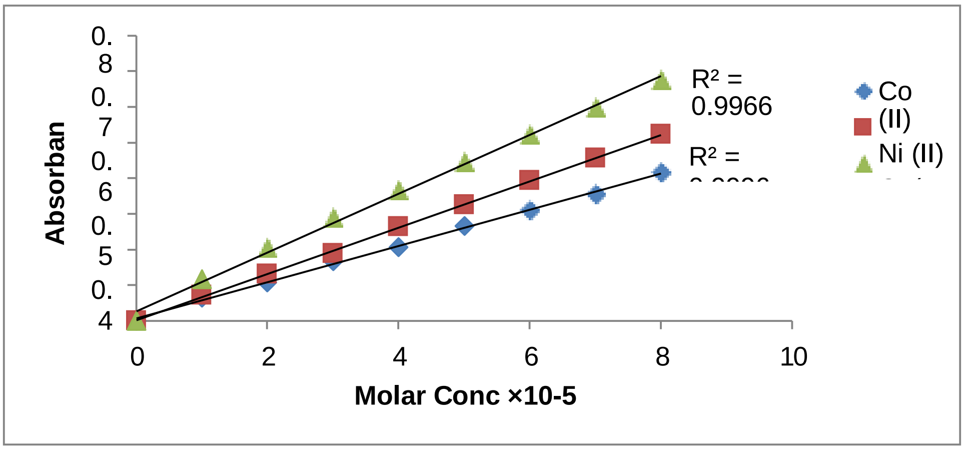
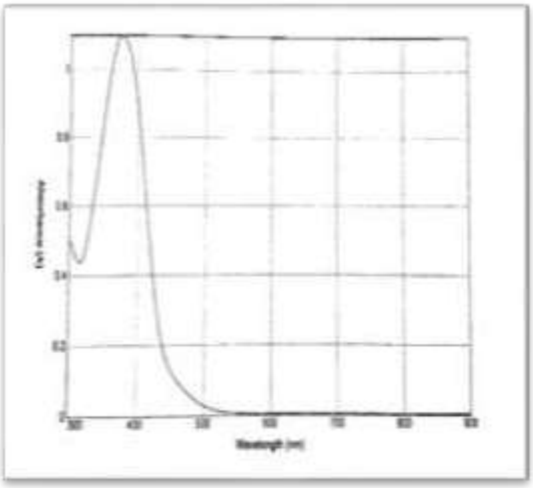


Fig. 5: Linear relationship between molar condensation and absorption

UV-Vis Spectral Studies

The UV-Vis spectra to the produced compounds melted at ethanol(10^{-4} M/L) were gauged as well the datum formed are listed at Table 2. A UV- Vis spectrum for the azo ligand shows peaks in 250 and 370 nm were appointed into mild energy (π-π*) transition and peak in 395 nm due to (n-π*) transition[16]. Co(II) spectrum appears three peaks at 242, 340 and 370 nm caused by intra ligand and a charge transfer, peaks at 420, 440 and 470 nm described to electronic transition type 4 T1g(F)→ 4 T1g(P), 4 T1g(F)→ 4 A2g and 4 T1g(F)→ 4 T2g(F) respectively [17]. Ni(II) complex exhibited three absorption peaks at 244, 355 and 388 nm which were assigned to intra ligand and charge transfer. Other peaks at 427, 445 and 488 nm were assigned to electronic transition type 3 A2g→ 3 T1g(P), 3 A2g → 3 T1g(F) and 3 A2g → 3 T2g(F) respectively [18]. Cu(II) complex shows peaks at 245, 353 and 425 nm due to intra ligand and charge transfer, while peak at 450 nm described to electronic transition type 2 Eg→2 T2g, which was very close to the octahedral environment[19]. 7739



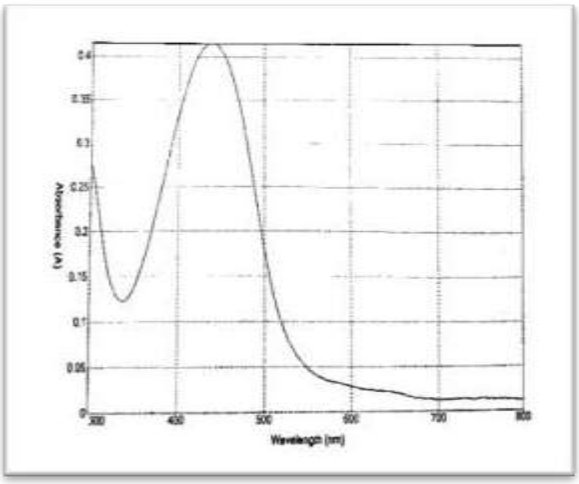
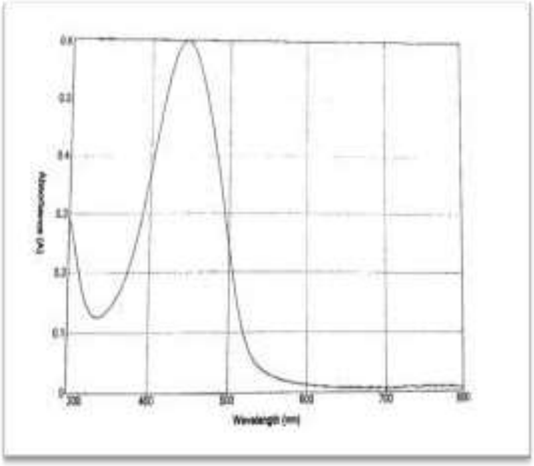


Fig. 6: UV-Vis spectrum of Ligand (SBPDBI) **Fig.7 : UV-Vis Spectrum for Cobalt(II) Complex With (SBPDBI)**



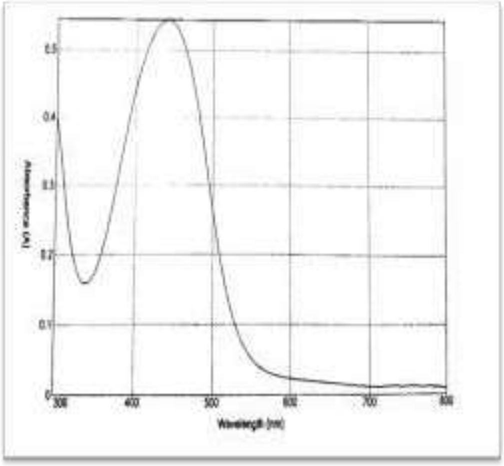


Fig.8 : UV-Vis Spectrum for Nickel(II)Complex With (SBPDBI) **Fig.9 : UV-Vis Spectrum for Copper(II)Complex With (SBPDBI)**

Table 2: Conditions of the produced compounds and UV- Visible, magnetic susceptibility as well as conductance mensuration's datum.

Compounds	(λmax)nm	M:L Ratio	Ɛ(L. mole^{-1} .cm^{-1})	(S.mol^{-1}.cm^{2}) in ethanol	Correlation Cofficient (R^2)
Ligand	390	------	------	-----	
$[Co_2L_2.4H_2O]Cl_4$	440	2:2	0.507×10^{3}	42.50	
$[Ni_2 L.4H_2O]Cl_4$	445	2:2	0.644×10^{3}	38.90	
$[Cu_2L_2.4H_2O]Cl_4$	450	2:2	0.81×10^{3}	42.08	

IR Spectra of Azo Ligands and Complexes

Infrared spectra The IR spectra data (KBr disk) of ligand (L) and Co(II), Ni(II) and Cu(II) complexes are summarized in Table 3.The IR spectrum of the free ligand (L) showed a medium and broad band around 3437.15 cm^{-1}, which can be attributed to (- N-H) stretching vibration of imidazole moiety [20-22]. The position of this band interacts with broad band at (3446-3454) cm^{-1} in the spectra of the complexes ,this an apparent change in the absorption values is due to of the hydrogen bonding between hydrogen of secondary amine in imidazole ring, in free ligand, and lone pair in an azo group[23]. The IR spectra of ligand appear band at (1629.85) cm^{-1} due to ν(C=N) of the N3 in the imidazole ring, which shifted to lower or the higher frequencies in the prepared complexes spectra due to the linkage of a metal ion with nitrogen imidazole ring [24,25].The ν(N=N) stretching vibration appears two bands at (1512 and 1440.83) cm^{-1} in the free ligand spectra, this bands shifted relatively to (1500-1402)cm^{-1} in the complexes spectra indicates the N=N group coordination. New weak bands in the region (439-470) cm^{-1} in the complexes spectra which were not present in the spectra of the free ligand may be attributed to ν(M-N) [26,27].

Table 3: Most Imperative FTIR spectral bands and their Assignments of the azo ligand and its Complexes (cm^{-1})

Compounds	υ (N-H)	υ (C=N)	υ (N=N)	υ(M-N
Ligand	3437.15	1629.85	1440.83	--------
[Co2 L2.4H2O]Cl4	3446.79	1629.85	1456.26	439.77
[Ni2 L.4H2O]Cl4	3448.72	1627.92	1458.18	468.70
[Cu2 L2.4H2O] Cl4	3454.51	1627.92	1458.18	459.06

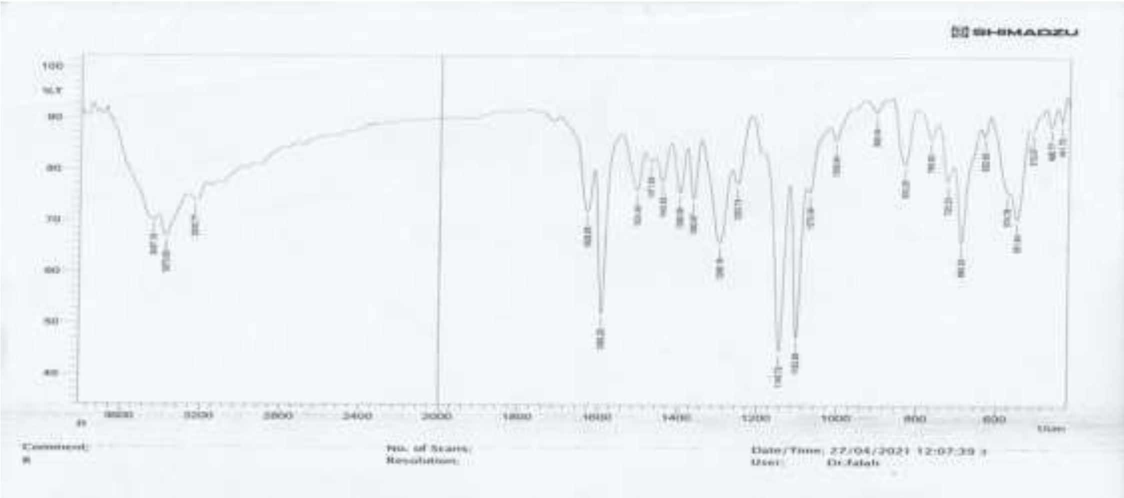


Fig. 10:FTIR spectrum of azo ligand (SBPDBI)

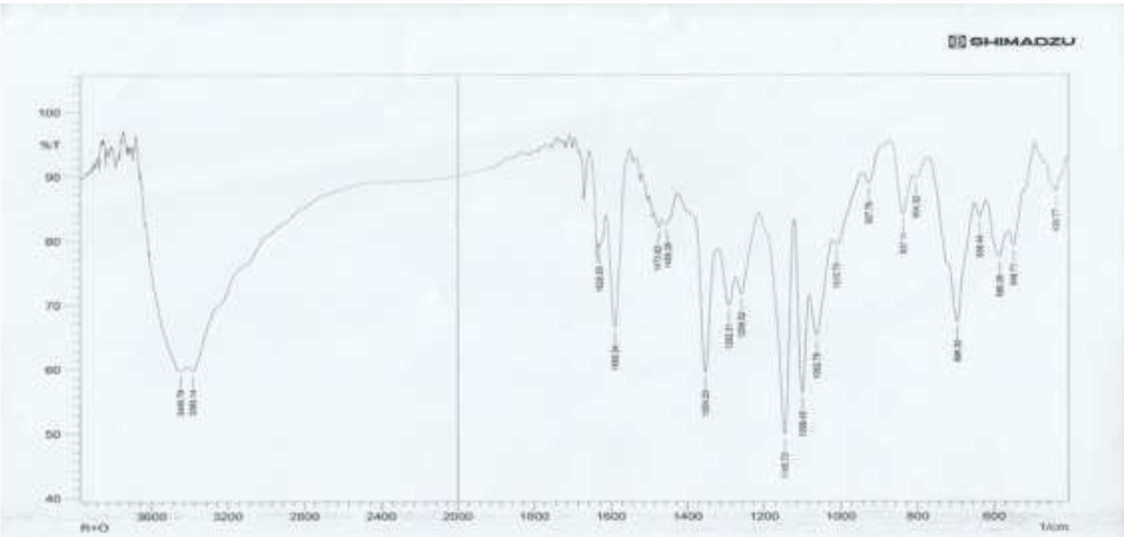


Fig.11 : **Infrared Spectrum of Co(II) Complex With (SBPDBI)**

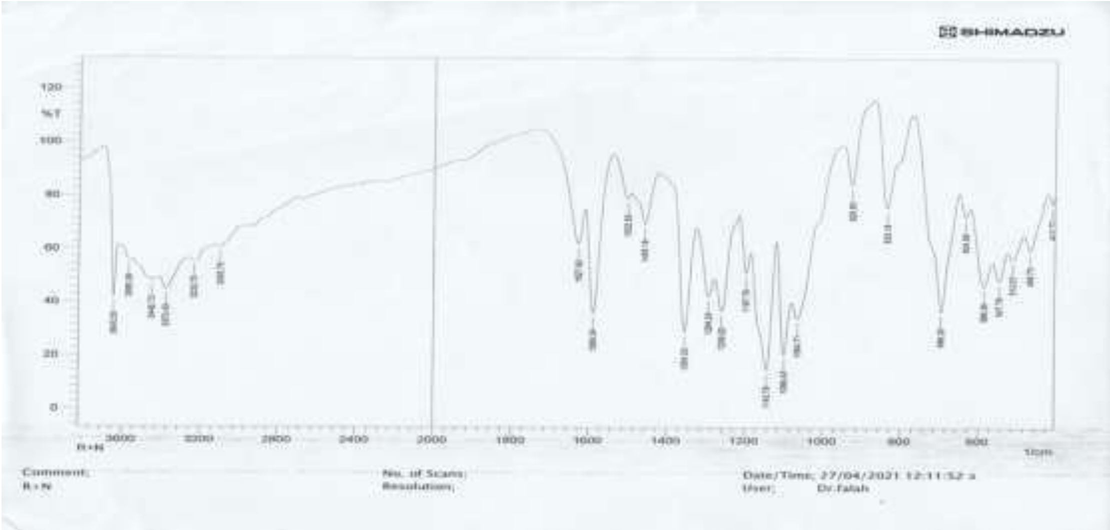


Fig.12: **Infrared Spectrum of Ni(II) Complex With (SBPDBI) .**

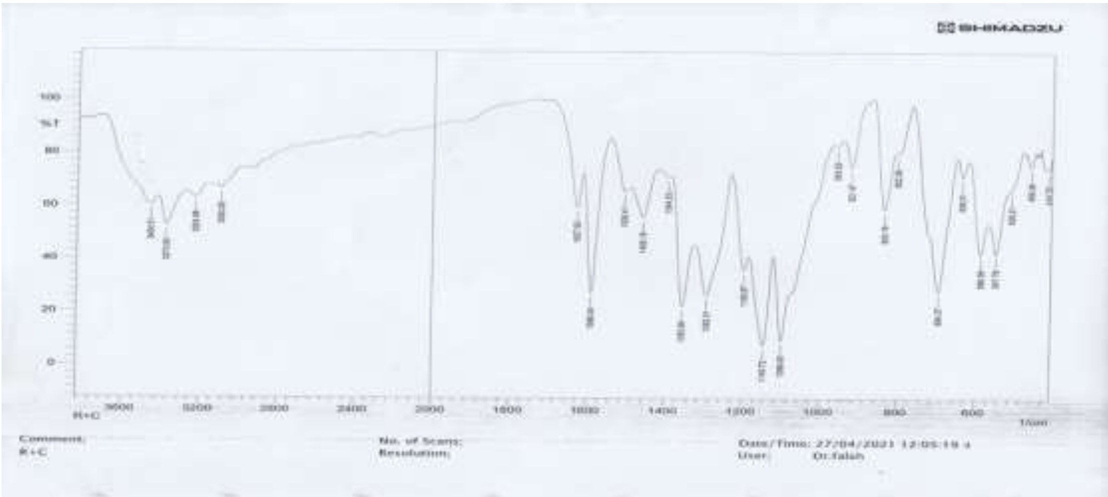


Fig.13 : **Infrared Spectrum of Cu(II) Complex With (SBPDBI)**

Antimicrobial Activity

The response of selected bacteria, we observed the excessive biochemical effect of the azo ligand and its complexes studied at concentration (1x10-3 M) with dual varieties of pathogenic bacteria (Staphylococcus aureus), its Gram Positive and either with the germ (Escherichia coli), Gram negative bacteria, have depicted lower response to ligand besides its complexes investigated from the other category of bacteria, categorized it's resistant to numerous chemical compounds and antibiotics[28],[29],[30]. The cause for this resistant is the colon bacteria that are found in a distinct bacilli having thick casing surroundings of its cell, this casing has an excessive amount of lipid works to oppose these materials from inflowing a cell, whereas the Staphylococcus aureus bacteria don't have this feature. Accordingly, it will be in lower resistance in an arrival of chemical and antibiotic substances to inner bacterial cell [31-33]. Normally, the prepared complexes have explained biochemical effect more as compared with the ligand (SBPDBI), while the ligand contain nitrogen and oxygen atoms biological retardant[34],[32],[35]. The positive charge ion in the chelated complex has been partly shared with the donor atoms orbital in a ligand and there has been π-electron delocali-zation over the entire chelate ring, which decrease in the polarity of the metal ion to a greater level, this in turn increases a lipophilic character of a metal chelate and favors its permeation via the lipid layers of the membrane the microorganisms[32],[35]. The consequences presented in previous activity Tables (5) and figures (14,15).

Table 5. The data of antibacterial activity (zone of inhibition) (mm) of azo ligand (SBPDBI) and its complexes.

Compound	G-(E.coli)	G+(St.aureus)
Control (S) DMSO	6	6
SBPDBI	15	18
[Co_2 L_2.4H_2O] Cl_4	17	20
[Ni_2 L_2.4H_2O] Cl_4	19	22
[Cu_2 L_2.4H_2O] Cl_4	17	21

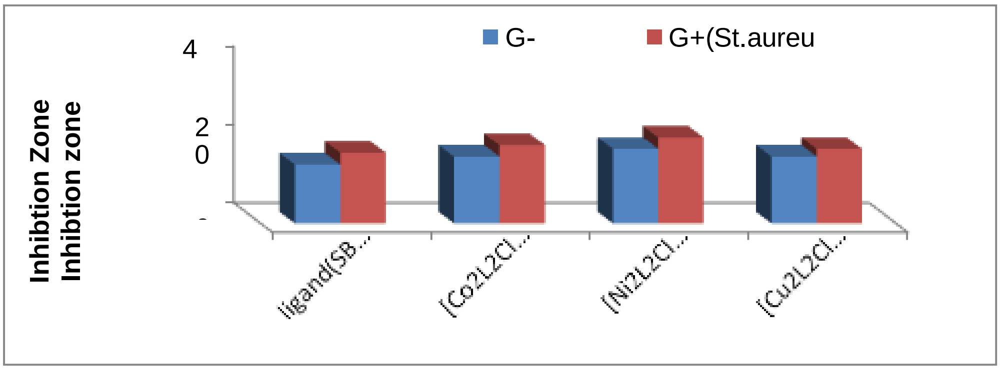


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Fig. 14: The biological effects of azo ligand (SBPDBI) and its complexes

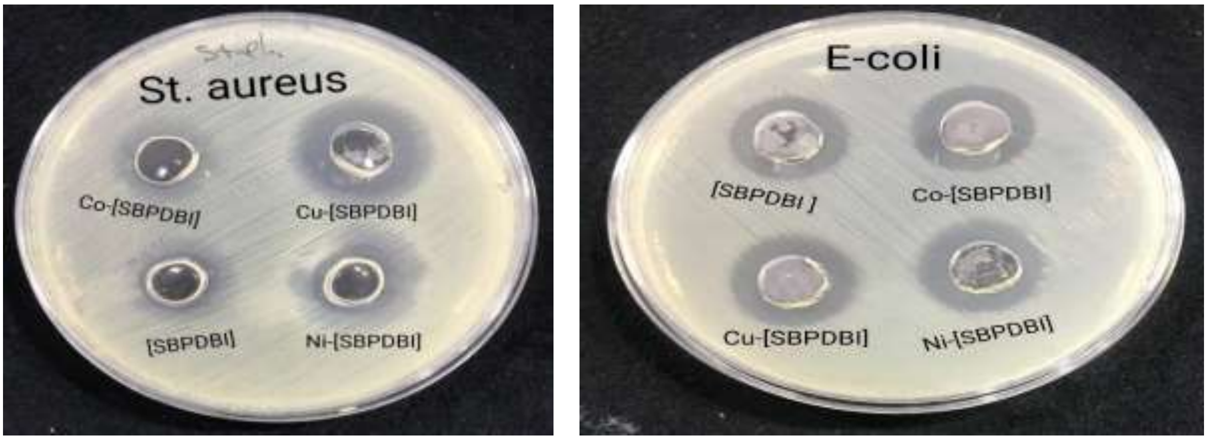

Fig. 15: Photograph of antibacterial activities of the ligand (SBPDBI) and its complexes

Conclusion

In this paper, we reported the synthesis and spectral characterization of new azo dye ligand derived from 4,4′-sulfonyl dianiline with imidazole (SBPDBI) and its metal complexes with Co (III), Ni (II) and Cu (II) ions. Further, a series of metal complexes comprising the ligand have been prepared and characterized by FT-IR, UV-Visb spectral studies. One the basis of their analytical and spectral data. This behavior with M:L mole ratio of 2:2. Each complex has been of octahedral geometry and categorized by the numerous physio-chemical techniques, the analysis ratified the composition and structures for the gained complex jointly. Biological activity results have depicted that all the compounds have variety of antibacterial activities.

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