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Synthesis, Characterization and Antibacterial Studies of Some Transition Metal Complexes Derived from 1-((2-Hydroxyphenyl)Iminomethyl) Naphthalene-2-ol

Lirwanu S, H. Anas, U. Ziyaulhaq

Lecturer II, Department of Chemistry, Al-Qalam University, P.M.B. 2137, Katsina, Nigeria
Assistant Lecturer, Department of Chemistry, Federal University, P. M. B. 1001, Gusau, Nigeria
Lecturer II, Department of Chemistry, Al-Qalam University, P.M.B. 2137, Katsina, Nigeria

ABSTRACT: Schiff base derived from the reaction of 2-amino phenol and 2-hydroxy-1-naphthaldehyde and its Mn(II), Co(II), Ni(II), Cu (II) and Zn (II) complexes has been synthesized. The Schiff base and its complexes were characterized by Solubility Test, Melting Point, Decomposition Temperature, Molar Conductance, IR and Magnetic Susceptibility. The number of ligands coordinated to the metal ion was determined using Job's Method of Continuous Variation. The ligand is orange (in color) with melting point temperature of 186^0C and percentage yield of 82%. The percentage yields of the complexes range from 51-75%. The decomposition temperatures of the complexes were in the range of 192-286^0C. Their molar conductance values (5.37-8.40 Ohm^{-1} cm^2mol^{-1}) indicate that all the complexes are non-electrolytes. Magnetic moment values of the complexes showed that Mn (II), Co (II) and Cu (II) are paramagnetic where as Zn (II) and Ni (II) are diamagnetic. The ligand and its metal (II) complexes were screened for antibacterial activity against *Staphylococcus aureus*, *Escherichia coli*, *Salmonella typhi* and *Streptococcus pneumoniae* using disc diffusion method. The results indicated that the ligand and its metal (II) complexes have activity at different concentrations against all the species of bacteria used.

KEYWORDS: Synthesis, Schiff base, Ligand, Complex, Characterization, Antibacterial activity

I. INTRODUCTION

The chemitry of Schiff base complexes is fast developing, especially those involving aldehydes and amines. This is because of the wide variety of possible structure of ligands [1]. Schiff bases have played an important role in the development of coordination chemistry as they readily form stable complexes with most of the transition metals. In the area of bioinorganic chemistry interest in Schiff base complexes has centered on the role of such complexes in providing synthetic models for the metal containing sites in metalloproteins and enzymes [2]. However, the incorporation of transition metal ions into the Schiff base have wide application in the field of analytical chemistry, food industry, dye industry, catalysis, fungicidal, agrochemical along with biological activities [3].

A wide variety of antibiotics have been developed against bacterial infections. Because of extensive overuse or misuse of antibiotics, bacteria are becoming antibiotics resistance resulting in growing threat to human health. This causes increase in mortality [4]. The need for effective therapies against multi drugs resistance bacteria has stimulated researchers into design and synthesis of novel antibacterial molecules [5]. Owing to this increased microbial resistance, new classes of antimicrobial agents with novel mechanisms are today's need to combat multidrug-resistant infections. Schiff base complexes of transition metals containing ligand with nitrogen and oxygen donors are known to exhibit interesting antibacterial activity [6]. From a report, synthesis, characterization and antimicrobial activity of Schiff base complex of Mn(II), Cu(II), Ni(II), Co(II) and Zn(II) derived from 3-aminophenol and 2-hydroxy-1-naphthaldehyde was studied. The Schiff base ligand and the metal complex were screened for their antimicrobial activity on different species of pathogenic bacteria and fungi. The result of these studies revealed that the complexes exhibit more activity against the various species compared to the Schiff base [7].

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The prime aim of this work is to synthesize, characterize and study the antibacterial activity of Schiff base derived from the reaction 2-aminophenol and 2-hydroxy-1-naphthaldehyde and its Mn(II), Co(II), Ni(II), Cu(II) and Zn(II) complexes.

II. MATERIALS AND METHODS

2-hydroxy-1-napthaldehyde was of analytical grade and procured from Sigma-Aldrich. Other chemicals/reagents were of analytical grade and used without further purification. The metal salts such as Cu(II), Ni(II), Co(II), Mn(II) and Zn(II) chlorides were obtained from chemical store, Bayero University, Kano. Four pathogenic bacteria viz: *Escherichia coli, Staphylococcus aureus, Streptococcus species* and *Salmonella typhi were* collected from the Department of Microbiology, Bayero University, Kano. Nutrient agar was used as bacteriological media.

All weighing were carried out on electrical balance model H3OAR, melting point and decomposition temperatures were recorded using Gallenkamp melting point apparatus. Conductivity measurements were carried out using Jenway conductivity meter model 4010 in DMSO solvent. The infrared spectral analyses were recorded using Shimadzu FTIR 8400S model. The magnetic susceptibility values were obtained at room temperature using mk1 Sherewood Magnetic Susceptibility Meter. The *in vitro* antibacterial screening was performed by disc diffusion method at Department of Microbiology, Bayero University, Kano.

A. Preparation of the Schiff base, 1-((2-HydroxyPhenyl)Iminomethyl)Naphthalene-2-ol

0.01mol 2-aminophenol in 25cm^3 ethanol was slowly added to a solution of 0.01mole of 2 – hydroxy –1–naphthaldehyde in 25cm^{3}ethanol. The mixture was refluxed for two hours. The Schiff base that formed was filtered and washed with ethanol and dried over phosphorus pentoxide [8].

B. Preparation of Metal (II) Schiff Base Complexes

A general method has been used for the preparation of complexes using the reaction of metal salts and the corresponding Schiff-base in a molar ratio (1:2). An ethanolic solution of ligand (0.04 mol) and metal (II) chlorides (0.02 mol) both were mixed gently and refluxed for 2 h. The volume of the resulting solution was concentrated, by evaporating the solvent. The reaction mixture was cooled to room temperature which solidified on cooling. The solid thus obtained was filtered, washed thoroughly with ethanol to apparent dryness and dried over phosphorus pentoxide [9].

C. Determination of Metal to Ligand Ratio (Job's Method)

0.001M of the ligand and the metal (II) Chlorides in DMSO were prepared. The following ligand to metal salt ratio (ml); 1:15, 3:13, 5:11, 7:9, 9:7, 11:5, 13:3 and 15:1 were taken from ligand solution and each of the metal (II) chloride respectively. A total volume of 16ml was maintained (in the order) through the process and the mole fraction of the ligand was calculated in each mixture. The solution of the metal (II) chlorides were scan (as blank) to find the wave length λ_{max} for the particular metal ion before taking the absorbance values. The absorbance values were extrapolated against the mole fraction of the ligand and the number of coordinated ligand was determined using the relation below

$$n = \frac{Xi}{1-Xi}$$

Where n is the number of coordinating ligand and Xi is the mole fraction at maximum absorbance

D. Antibacterial Activity

The ligand and complexes were dissolved separately in DMSO to have four different concentrations (500µg 1000 µg, 2000µg, and 4000µg) per disc. They were placed on the surface of the culture and incubated at 37^{o}C for 24hrs. The diameter of the cleared zone of inhibition surrounding the disc is taken as the measure of inhibitory power of the sample against *Staphylococcus aureus*, *Escherichia coli*, *Salmonella typhi* and *Streptococcus pneumoniae* [10]. The antibacterial activity was carried out by disc diffusion method.

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III. RESULTS AND DISCUSSION

A. RESULTS

Table 1: Physical Properties of the Ligand and its Corresponding Metal (II) Complexes

Ligand/complexes	Colour	Yield (%)	Melting point (^{0}C)	Decomposition temperature (^{0}C)
Ligand	Orange	82	186	_
[MnL_2]	Cream	56		240
[CoL_2]	Brown	75		280
[NiL_2]	Dark yellow	51		192
[CuL_2]	Dark green	62		230
[ZnL_2]	Orange	68		286

Table 2: Solubility of the Ligand and its Metal (II) Complexes in some common solvents

Ligand/ Complex	Distilled Water	Methan ol	Ethanol	DMSO	DMF	$CHCl_3$	n-Hexane	CCl_4
Ligand	IS	S	S	S	S	S	SS	IS
[MnL_2]	IS	S	S	S	S	SS	SS	IS
[CoL_2]	IS	S	S	S	S	SS	SS	IS
[NiL_2]	IS	S	S	S	S	SS	SS	IS
[CuL_2]	IS	SS	SS	S	S	SS	SS	IS
[ZnL_2]	IS	S	S	S	S	SS	SS	IS

Key: S=Soluble, SS=Slightly Soluble, IS=Insoluble, L=Ligand

Table 3: The Infrared Frequencies of the Ligand and its Metal (II) Complexes.

Ligand/complex	ν (OH) cm^{-1}	ν (C=N) cm^{-1}	ν (C-O) cm^{-1}	ν (M-O) cm^{-1}	ν (M-N) cm^{-1}
Ligand	3119	1618	1356	_	_
[MnL_2]	3440	1627	1428	528	393
[CoL_2]	3446	1628	1438	538	406
[NiL_2]	3396	1619	1412	517	413
[CuL_2]	3401	1608	1396	537	413
[ZnL_2]	3438	1625	1428	533	459

Table 4: Molar Conductance of the Complexes in 10^{-3}M Solution of DMSO

Complex	Concentration in moldm^{-3}	Specific Conductance Ohm^{-1} cm^{-1}	Molar Conductance Ohm^{-1} cm^{2}mol^{-1}
[MnL_2]	1×10^{-3}	8.40×10^{-6}	8.40
[CoL_2]	1×10^{-3}	7.07×10^{-6}	7.07
[NiL_2]	1×10^{-3}	7.53×10^{-6}	7.53
[CuL_2]	1×10^{-3}	5.83×10^{-6}	5.37
[ZnL_2]	1×10^{-3}	5.37×10^{-6}	5.83

Table 5: Magnetic Susceptibility Measurement

Compound	Gram Susceptibility, χ_g (erg•G^{-2} g^{-1})	Molar Magnetic Susceptibility , χ_M (erg•G^{-2}/ mol)	Effective Magnetic Moment in Bohr Magnetons (μ_{eef})
[MnL_2]	3.26×10^{-6}	1.88×10^{-3}	2.11
[CoL_2]	7.5×10^{-6}	4.37×10^{-3}	3.21
[NiL_2]	$- 6.44 \times 10^{-7}$	-3.75×10^{-4}	Dia
[CuL_2]	2.53×10^{-6}	1.48×10^{-3}	1.87
[ZnL_2]	-4.3×10^{-7}	-2.5×10^{-4}	dia

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Table 6: Antibacterial Screening of the Ligand and its Corresponding Metal (II) Complexes

Test Organism	Ligand/Complex	Zone of Inhibition(mm)/Concentration in(μg)			
		4000	2000	1000	500
Staphylococcus Aureus	Ligand	10	08	07	06
	$[MnL_2]$	10	08	07	06
	$[CoL_2]$	12	09	06	06
	$[NiL_2]$	06	06	06	06
	$[CuL_2]$	10	08	06	06
	$[ZnL_2]$	06	06	06	06
	Ciprofloxacin	23	20	17	15
Escherichia coli	Ligand	11	09	07	06
	$[MnL_2]$	06	06	06	06
	$[CoL_2]$	16	12	08	06
	$[NiL_2]$	12	10	08	06
	$[CuL_2]$	10	07	06	06
	$[ZnL_2]$	08	06	06	06
	Ciprofloxacin	24	20	14	10
Streptococcus pneumonia	Ligand	12	10	08	06
	$[MnL_2]$	16	12	10	08
	$[CoL_2]$	12	09	07	06
	$[NiL_2]$	13	10	09	07
	$[CuL_2]$	15	12	08	06
	$[ZnL_2]$	14	12	10	06
	Ciprofloxacin	25	21	18	16
Salmonella typhi	Ligand	06	06	06	06
	$[MnL_2]$	06	06	06	06
	$[CoL_2]$	06	06	06	06
	$[NiL_2]$	06	06	06	06
	$[CuL_2]$	08	06	06	06
	$[ZnL_2]$	06	06	06	06
	Ciprofloxacin	20	16	14	10

B. DISCUSSION

From Table 1, the Schiff base derived by the condensation of 2-amino phenol and 2-hydroxy-1-naphthaldehyde was orange in color, stable and non hygroscopic solid at room temperature. The corresponding product was obtained in good yield (82%). The melting point of the ligand was 186^0C. The reaction of ethanolic solutions of the Schiff base and the respective metal (II) chlorides yield 51-75% of the metal (II) Schiff base complexes. The colors of Mn (II), Co (II), Ni (II), Cu (II) and Zn (II) complexes are cream, brown dark yellow, dark green and orange respectively. The decomposition temperatures (Table 1) of these complexes were in the range of 192-286^{o}C. The value of melting point and that of the decomposition temperatures indicate that the complexes are stable [11].

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The solubility tests (Table 2) of the Schiff base ligand and its metal (II) complex in water and common organic solvents carried out revealed that the ligand and its Mn (II), Co (II), Ni (II), Cu (II) and Zn (II) complexes are all soluble in methanol, ethanol, DMSO and DMF. Cu (II) complex was found to be slightly soluble in methanol and ethanol. However, all the complexes were found to be insoluble in distilled water and tetrachloromethane and slightly soluble in chloroform and n-hexane. The ligand was found to be soluble in chloroform, slightly soluble in n-hexane and insoluble in distilled water and tetrachloromethane [12].

The IR spectral data of the ligand and its complexes are presented in Table 3. The nature and possible mode of bonding of the ligand with different metal ion have been studied by comparing the IR spectra of the ligand and that of complexes. The ligand exhibits bands at 3119 cm^{-1} due to ν (-OH) of aromatic. The band at 1618cm^{-1} is characteristic of the azomethine nitrogen atom in the free ligand. Evidence of nitrogen coordination of azomethine ν(C=N) group to the central metal atom from the shift of the frequency from 1618cm^{-1} in the ligand to 1608-1628cm^{-1} in all the metal complexes. This is further supported by the appearance of a new band at 393-459 cm^{-1} due to ν (M-N) bond. The band due to ν (M−O) stretching vibrations is observed at 517-538 cm^{-1} which support the involvement of oxygen atom in coordination. The band in the metal Schiff base complexes in the range of 3396-3446 cm^{-1} are due to *v* (O-H) stretching vibration [13]. The molar conductivity values for the complexes in DMSO were in the electrolytic range of 5.37-8.40 µs. Conductivity measurements (Table 4) have frequently been used in the structural elucidation of metal chelates (mode of coordination) within the limits of their solubility. They provide a method of testing the degree of ionization of the complexes, the molecular ions that a complex liberates in solution, the higher will be its molar conductivity and vice versa. It is clear from the conductivity data that the complexes present seem to be non electrolytes. Also the molar conductance values indicate that there is an absence of anions outside or inside the coordination sphere. Magnetic moment values (Table 5) of the complexes showed that Mn (II), Co (II) and Cu (II) are paramagnetic where as Zn (II) and Ni (II) are diamagnetic. The number of ligand coordinated to the metal ions was found to approximately be equal to 2 and the metal to ligand ratio was found to be 1: 2 [14].

The antibacterial activity test for the ligands and the complexes has been determined. The diameter of zone of inhibition (mm) was measured for each treatment. The ligands showed minimal activity against the entire organisms (Table 6). No activity recorded against *Salmonella typhi* by the ligand and the entire complexes with the exception of [CuL_2] which showed activity at higher concentration of 4000µg. The complexes showed activity on the isolates. However, *Staphylococcus aureus* is resistance to [NiL_2] and [ZnL_2] at all concentrations, but showed activity against the ligand [MnL_2] and highest activity was recorded at 4000µg against [CoL_2]. The complex [MnL_2] and [CoL_2] showed strong activity on the isolate. Highest zone of 16mm was recorded on *E.coli*, by [CoL_2] and *Streptococcus pneumoniae* by [MnL_2]. The ligand, [CuL_2] and [ZnL_2] showed activity at 4000µg, 2000µg and 1000µg against Escherichia *coli*, with no activity at 500µg and by [MnL_2]). Both the ligand and the complexes showed minimal activity when compared to the reference drug. The antibacterial activity of the ligand and its complexes were found to increase with increase in concentration, thereby indicating the important role of concentration in increasing the degree of inhibition [13].

From the analyses carried out, the structure of the ligand and that of the complexes were proposed as follows:

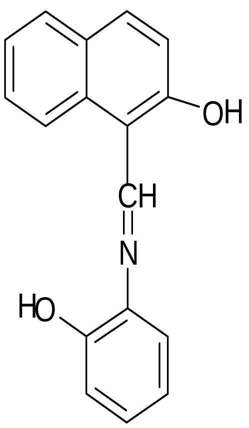

Fig. 2: Proposed Structure of the Ligand

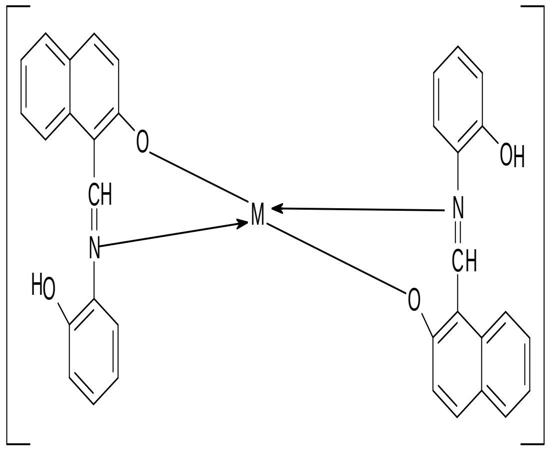

Fig. 1: Proposed Structure of the Complexes

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IV. CONCLUSION

This research effort examined the synthesis, characterization and antibacterial activity of novel coordination complexes of some metals with a Schiff base derived from 2-aminophenol and 2-hydrxy-1-naphthaldehyde. *In vitro* antibacterial activity of these compounds was tested by using Disk Diffusion Method, in which *Staphylococcus Aureus*, *Eschericia coli*, *Streptococcus pneumonia* and *Salmonella typhi* were employed as test organisms. The results indicated that both the ligand and the metal (II) complexes showed activity on the isolates at some concentrations and showed no activity on the isolates at other concentrations.

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