



Original Research Article

**Nematicidal Activity Studies of Cu<sup>2+</sup> and Zn<sup>2+</sup> Complexes with Some Aldimine Ligands Against *Meloidogyne arenaria*: A Root Knot Nematode of *Arachis hypogea***

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**ABSTRACT**

Two Schiff base donor ligands (aldimines) were used to prepare four transition metal complexes of Cu(II) and Zn(II) by treating 2-nitroaniline and salicylaldehyde to form the Schiff base ligand 2-[(2-nitrophenylimino) methyl] phenol (L<sub>1</sub>) and 4-nitroaniline and salicylaldehyde to form 2-[(4-nitrophenylimino)]methyl]phenol (the L<sub>2</sub> counterpart). The processes were accomplished using a microwave oven assisted reaction regulated at 100°C for 30 min. Characterization was carried out on the basis of physical properties elemental analysis, FTIR, <sup>1</sup>H-NMR and <sup>13</sup>C-NMR spectroscopy, electronic spectral and mole ratio method. Results from the FTIR spectra of the ligands and corresponding complexes showed that the ligands L<sub>1</sub> and L<sub>2</sub> displayed a bidentate character with coordination via the nitrogen and oxygen atoms. The <sup>1</sup>H-NMR and <sup>13</sup>C-NMR data of L<sub>1</sub> and L<sub>2</sub> confirmed the formation of the complexes of Cu(II) and Zn(II) showing that the probable coordination geometries of Cu(II) and Zn(II) are octahedral. The complexes were found to be non electrolytes in alcohol. The nematicidal activity studies implied that the ligands and their metal complexes showed promising nematicidal ability with the metal complexes showing comparatively higher inhibiting ability than the free ligands against the root knot nematode *Meloidogyne arenaria* present in the crop *Arachis hypogea*.

**Keywords:** Nematicidal activity, Aldimines, *Meloidogyne arenaria*, *Arachis hypogea*

## Introduction

The interaction of metal ions with organic ligands shows better antimicrobial activity compared to free ligands (not coordinated), and as such, it justifies the investigation of new drugs with unknown mechanism of action against pathogenic bacteria. The use of these novel compounds is likely to have great potency against pathogens; albeit, the quest for nascent ideas of evaluating the antimicrobial activity cannot be overemphasized [1-4]. It is a major fact that, lipophilicity of a drug is enhanced through chelate formation which raises the drug activity due to its ease of permeability into the active site. The central metal atoms/ ions play a very important function in numerous biological processes; ions with biologically active ligands are substances of reasonable interest. Some of the biological active compounds like azines act through chelation process but for most of them only small data are available about how metals binding influence their ability. Some heterocyclic azines are known to inhibit the development of marine tumor and serve as fluorescent brightening agent and are photo-sensitizer. Azines have been produced for ion selective optical sensors. Mixed azines between opioid antagonist and steroidal ketones have been shown to possess various metal complexes so are their antimicrobial and antifungal properties [3-6]. Detection of metal ions of biological importance has attracted much attention. Like  $Zn^{2+}$  ion fluorescent probe sensors have achieved special interest.  $Zn^{2+}$  is an essential trace element and the second (after  $Cu^{2+}$ ) most abundant metal ion in humans [7, 8]. Metal complexes of aldimines and ketimines have contributed significantly to the recent advancements in coordination and bioinorganic chemistry. These Schiff bases present an array of diverse and flexible categories of ligands capable of coordinating with different metal ions to give complexes with the right chemical properties for suitable applications. In recent times, focus has been shifted to the chemistry of the metal complexes of Schiff bases which possess nitrogen, oxygen and other donor atoms. This may be

attributed to their application in many fields [4, 9, 10]. Various studies have shown that the metal complexes have a higher activity against micro-organisms compared to the free ligands because of chelation [11-13]. This increased movement is due to the fact that the chelation tends to reduce the metals polarity. This reduced polarity then increases the lipophilic properties of the metals. This eventually favours the penetration of the complexes across the membrane and blocks the active sites which the microbes are supposed to act. [14, 15]. Schiff bases are the compounds containing azomethine group ( $-\text{HC}=\text{N}-$ ). They are condensation products of ketones (ketimines) or aldehydes (aldimines) with primary amines and were first reported by Hugo Schiff in 1864 [16]. Formation of these Schiff bases generally occurs under acids or base catalysis or increased temperature, i.e. by heating the reaction process. The common Schiff bases are crystalline solids, which are slightly basic but at least some of the Schiff bases form insoluble salts with strong acids. Schiff bases are used as intermediates for the synthesis of amino acids or as ligands for preparation of metal complexes having a series of different structures [17]. The resistance of nematodes is fast becoming a global concern due to the rapid increase in multidrug resistance microorganisms. As a result of the resistance of nematodes, some drugs that were previously used in the treatment of certain diseases are no longer effective. This has led to the quest for new drugs with relatively higher efficiency that can fit into the binding sites of target molecules [18-19].

In synthetic drug design, advantage has been harnessed from the knowledge of active site of targeted amino acids which are associated with a particular disease in order to combat the disease. Thus drug modeling begins from understanding the three dimensional build up of a protein so as to produce drugs that will complement the target enzyme in terms of stereochemistry and pattern of physiochemical activities thereby inhibiting normal chemical action and ultimately halting their progress [20]. This rational design process is accompanied by determination of the three

dimensional build up, production of modified compounds, synthesis and analyzing the new drugs prepared [21-22]. A great deal of work has been reported on the synthesis, structural investigations, various crystallographic features, mesogenic characteristics and structure-redox relationship and catalytic properties of different type of Schiff bases and their complexes with transition and non-transition elements, this was done in order to provide database base on the structure and chemical properties of the various Schiff base and their metal complexes but few data is available on the synthesis, Characterization and nematicidal properties of 2-[(2-nitrophenylimino) methyl] phenol and 2-[(4-nitrophenyliminomethyl)] phenol Schiff base ligands and their metal complexes as data obtained from such might assist in agriculture [3, 14, 24]. This work is aimed at synthesizing, characterizing and investigating the nematicidal properties of 2-[(2-nitrophenylimino) methyl] phenol and 2-[(4-nitrophenyliminomethyl)] phenol schiff base ligands and their complexes with Cu(II) and Zn(II) against the root knot nematode *Meloidogyne arenaria* present in the crop *Arachis Hypogea*.

## Materials and Methods

### Apparatus

The apparatus include: Boiling tubes, conical flask, spatula, filter paper spatula, wire gauze, microwave oven, funnels, beakers, refrigerator, thermometer 5mL syringe and tweezers.

### Reagents

Copper (II) chloride, Zinc (II) chloride, Salicylaldehyde, 2-nitroaniline, 4-nitroaniline, Ethanol, Distilled water among others were the reagents used for the experiment. Analytical grade reagents were used for the preparation and they were of high percent purity.

### **Preparation of the Ligand 2[(2-nitrophenylimino) methyl] phenol (L<sub>1</sub>)**

The method of preparing the aldimine ligand L<sub>1</sub> was done in accordance with the method described elsewhere [25]. Exactly 5.0g (0.01 mmol) of 2-nitroaniline accurately weighed into a crucible and pulverized into fine powder and 5mL of salicylaldehyde and 20mL of absolute ethanol added. The entire mixture was stirred for 1h and placed in a thermostated microwave oven DHG – 9101-1SA PEC Medical USA model and power source of 220V for 30 minutes. Stirring was continued and the temperature of the oven was maintained at 100°C. A thermocouple device was used to monitor the temperature and using on/off cycling to control temperature. The bright yellow solid product formed was filtered, washed with distilled water, air dried for 30 minutes and preserved in a refrigerator.

### **Preparation of the Ligand (L<sub>2</sub>) 2-[(4-nitrophenylimino)]methyl]phenol**

The aldimine ligand (L<sub>2</sub>) 2-[(4-nitrophenylimino)methyl]phenol] was prepared in accordance with a method described by [25] with slight modifications, 4-nitroaniline was treated with salicylaldehyde instead of 2-nitroaniline. The black solid product formed was filtered, washed with distilled water and air-dried.

### **Synthesis of metal complexes**

Using a 250 mL beaker, 5.0 (0.01 mol) of Schiff base L<sub>1</sub> and 20 mL ethanol was added drop wise into 1.0 mmol of metal salts of Cu (II) chloride and Zn (II) chloride, respectively. Each mixture was stirred thoroughly and placed into a microwave oven for 30 minutes. Each metal complexes formed was filtered, washed with distilled water. Where the product was oily, 10 mL ethanol was added to the mixture, stirred and placed into a microwave. The final product formed was air-dried

and preserved in a refrigerator at - 10° C. The same method was adopted for the synthesis of the metal complexes of the second ligand L<sub>2</sub>.

## **Determination of physicochemical properties of L<sub>1</sub>, L<sub>2</sub> and its complexes**

### **Melting point**

A small amount of the sample was introduced into a capillary tube which was attached to the stem of a thermometer centred in a heating bath while observing the temperatures at which melting begins and completes.

### **UV-Vis Spectra**

The UV – Vis spectra were recorded on a Shimadzu UV 160 I PC UV – VIS Spectrophotometer at the Federal University of Agriculture, Makurdi, Nigeria.

## **Nematicidal Studies**

### **Sampling Location**

The study was carried out on a farm land by the North Bank flank of the River Benue, Nigeria. No specific permits were required for the farm land needed for the studies and the sites were unprotected. The sites did not contain any endangered species of crops or animals. The test crop is peanut (*Arachis hypogea*) and the test nematode is *M. arenaria*.

### **Isolation and Identification of *M. arenaria***

Clean Tweezers were used to collect adult of *M. arenaria* suspect from galls of peanuts. After cleaning the galls with de-ionized water, the tweezers were used to scratch the root nodules of the

*Arachis hypogea* so as to expose the mature *M. arenaria*. The crop was identified in the Department of Botany while the nematodes were identified in the Department of Zoology, Federal University of Agriculture, Makurdi, Nigeria. The isolated nematodes, were kept in plates (one in each labeled appropriately) with 5 mL water. The aldimine ligands L<sub>1</sub> and L<sub>2</sub> and their metal complexes were prepared at the Research Laboratory Chemistry Department, U.A.M. The compounds were soaked in distilled water at different concentrations of 125, 62.5, 31.3, 15.6 and 7.8 ppm for 1 week. After filtration, 10mL of the extracts with various concentrations were transferred to 10 petri dishes in order to study the nematicidal effects. An equal volume of distilled water was used as a control. The soil with serious root nematode diseases was considered for the experiment.

### **Application of synthesized ligands and metal complexes on *M. arenaria***

The synthesized ligands and metal complexes were applied to the nematodes and observed for 30 minutes. The corrected percentage mortality was calculated. The nematodes, *M. arenaria* were considered alive if they were mobile or appeared as a winding shape. The nematodes were considered dead if they did not move when probed with a needle. To check if nematodes regain mortality or not they were transferred to distilled water for 12 h.

The corrected percentage mortality was calculated according to Equation (1) below:

$$\% \text{mortality} = \frac{(\text{mortality of treatment} - \text{mortality of } C_0)}{(1 - \text{mortality of } C_0)} \times 100 \quad (1)$$

Where C<sub>0</sub> = zero concentration of ligands and complexes

## Results and Discussion

### Analytical Data and Physical Property of L<sub>1</sub>, L<sub>2</sub> and its metal complexes with their conductivity values

Table 1 below presents the analytical data and physicochemical properties of the ligands L<sub>1</sub>, L<sub>2</sub> and their metal complexes.

**Table 1:** Elemental Analysis of L<sub>1</sub>, L<sub>2</sub> and its Metal Complexes

EMPIRICAL FORMULARS/ MOLAR MASS (g/mol)	Elemental Analysis calculated(found)						COLOUR	MPK	YIELD %	Ω M <sub>2</sub>	Magnetic moment BM
	C%	H%	N%	O%	M%	U%					
C <sub>13</sub> H <sub>10</sub> ON <sub>2</sub> O <sub>4</sub> (HL <sub>1</sub> ) (258)	65.1	4.41	14.2	10.8	-	-	Bright yellow	-	-	11.5	-
C <sub>13</sub> H <sub>10</sub> N <sub>2</sub> O <sub>3</sub> (HL <sub>2</sub> ) (258)	(64.9)	(4.9)	14.1	10.7			Dark brown				
[CuL <sub>1</sub> CL <sub>2</sub> ].4H <sub>2</sub> O(465)	48.2	4.3	10.6	8.0	8.0	8.9	Pale blue	1357	80.0	14.3	1.84
[CuL <sub>2</sub> Cl <sub>2</sub> ].4H <sub>2</sub> O(465)	48.0	4.2	10.5	7.9	7.9	8.9	Pale green				
[ZnLiCl <sub>2</sub> ].4H <sub>2</sub> O(465.4)	51.6	3.8	11.3	8.6	8.8	9.5	Dark maron	1180	86.0	9.9	Dia
[ZnL <sub>2</sub> Cl <sub>2</sub> ].4H <sub>2</sub> O(465.4)	51.5	3.7	(11.2)	(8.5)	(8.7)	(9.4)	Reddish	1180			

**Table 2:** Electronic Spectra of L<sub>1</sub>, L<sub>2</sub> and its metal complexes

Synthesized ligands and metal complexes	λnm	νcm <sup>-1</sup>	Transition	Suggested structure
1. [CuL <sub>2</sub> Cl <sub>2</sub> ].4H <sub>2</sub> O	248	40,487	Ligand field	
	271	36,766	Ligand field	
	390	25,533	Crystal field	
	660	15,121		- Octahedral
	487	20,566		
	415	24,151		
4. [Zn[L <sub>2</sub> Cl <sub>2</sub> ].4H <sub>2</sub> O	245	40,816	Ligand field	
	282	35,587	Ligand field	Octahedral
	36.7	27,321	Crystal field	



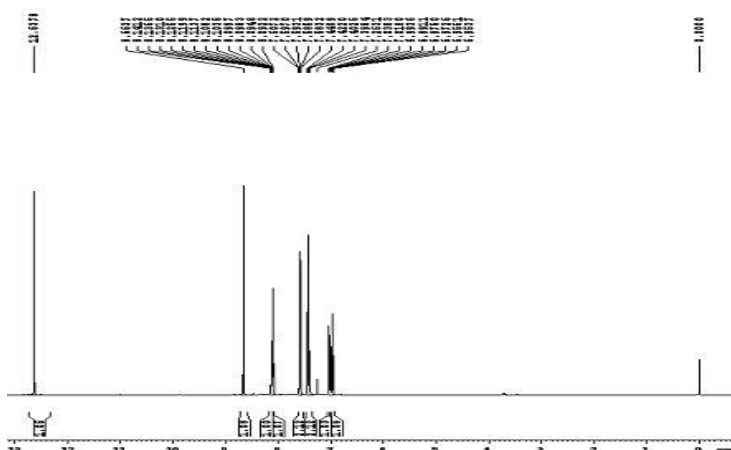
## Electronic Spectral studies

The ligand ( $L_1$ ) gave a sharp band at 229 nm corresponding to  $\pi - \pi^*$  transition with an additional sharp band at 230 nm. A broad band appeared at 340 nm corresponding to  $n - \pi^*$  involving molecular orbital of carbonitrile chromophore and aromatic ring while the ligand ( $L_2$ ) showed two absorption bands corresponding to  $\pi - \pi^*$  transition. At lower energy,  $L_1$  equally showed sharp bands while other bands occurred after 340 nm. A wavelength of 352 nm recorded  $n - \pi^*$  transition. On the other hand,  $\pi - \pi^*$  remained unchanged in the metal complexes but an increase in the Energy (E) value was observed. Metal complexes with the ligand in  $\pi - \pi^*$  transition showed that there was an enhancement in transition. The ligand ( $L_1$ ) gave a sharp band at 229 nm corresponding to  $\pi - \pi^*$  transition with an additional sharp band at 230 nm. A broad band appeared at 340 nm corresponding to  $n - \pi^*$  involving molecular orbitals of carbonitrile chromophore and aromatic ring. The ligand ( $L_2$ ) showed two absorption bands corresponding to  $\pi - \pi^*$  transition. At lower energy,  $L_1$  equally showed sharp bands. Other bands occurred after 340 nm. At 352 nm an  $n - \pi^*$  transition appeared. On the other hand  $\pi - \pi^*$  remained unchanged in the metal complexes but an increase in the E value was observed. Upon complexation of the metal with the ligand, there was an improvement in  $\pi - \pi^*$  transition. The metal complex,  $[CuL_1CL_2].4H_2O$  showed a sharp band at 280 nm. A broad band appeared at 344 nm. Metal complex  $[CuL_2CL_2].4H_2O$  showed twin bands at 320 – 370 nm. The UV-visible absorption spectra of all the metal complexes show similarities, which indicates similarity in their structures and generally showed the characteristic bands of the free ligands with some changes both in frequencies and intensities. During complex formation, the absorption bands of the complexes to some extent shifted to shorter wavelength (Blue shift) compared to those of the ligand. These adjustments of

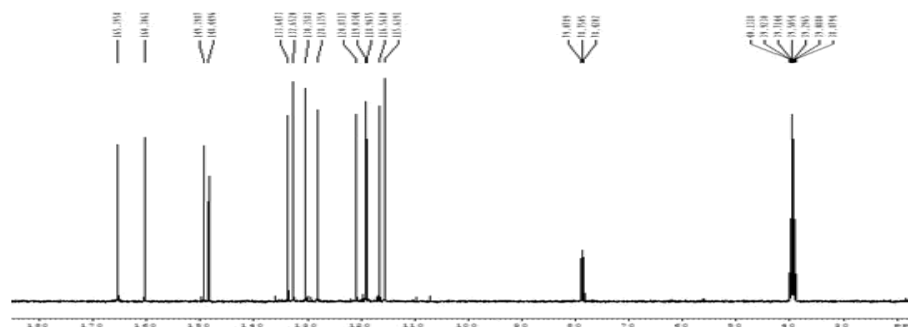
the shifts and intensity of the absorption bands indicated the coordination of the ligand to the metal ion.

### <sup>1</sup>H-NMR Data

The <sup>1</sup>H-NMR spectra of the synthesized compounds were recorded on 300 MHz NMR spectrometer. The resonance peaks in the <sup>1</sup>H NMR spectra of the ligands L<sub>1</sub> and L<sub>2</sub> were recorded in CdCl<sub>3</sub> and the synthesized metal complexes in dimethylsulfoxide (DMSO). The resultant resonance which was assigned by intensity showed promising composition of the compounds. In L<sub>1</sub> and L<sub>2</sub>, the hydroxyl proton displayed signals at 15.20 and 7.8 ppm respectively, which later disappeared in all the synthesized complexes due to the deprotonation of the hydroxyl proton. For the aromatic protons, L<sub>1</sub> and L<sub>2</sub> showed a simple pattern which is due to the presence of fewer protons in this region. The ratio of L to M was 2:1. The signals for 13 protons were observed after complex formation. The azomethine proton of L<sub>1</sub> and L<sub>2</sub> showed signals in the range 8.6 and 8.9 for L<sub>1</sub> and L<sub>2</sub>, respectively. A downwards shift of both the signals to 9.0 and 8.9 indicated a bonding of N to the metal.



**Figure 1a:** <sup>1</sup>H-NMR Spectrum for Schiff base 2[(2-nitrophenylimino) methyl] phenol (L<sub>1</sub>)

Figure 1b:  $^1\text{H}$ NMR Spectrum for Schiff base 2-[(4-nitrophenylimino)]methyl]phenol ( $L_2$ ) .**Table 3:**  $^1\text{H}$ NMR Spectral studies of  $L_1$  and  $L_2$  and its metal complexes

H-No	(1)	(5)	(7)	(11)
$L_1$	13.2(5)[2H]	12.7(5) [2H]	12.3(s)[2H]	15.0(s)[2H]
$L_2$	13.2(5) [2H]	12.7(5) [2H]	12.3 (s) 2H	12.75 (s) [2H]
$[\text{Cu}L_1\text{Cl}_2].4\text{H}_2\text{O}$	6.9-7.6(m) [7H]	7.0-7.8(m)[14H]	6.6-7.8m [7H]	6.8-7.5 (m) 14H
$[\text{Cu}L_2\text{Cl}_2].4\text{H}_2\text{O}$	6.9-7.6(m)[7H]	7.0-7.8(m)[14H]	6.6-7.8m [7H]	6.8-7.5 (m) [14H]
$[\text{Zn}L_1\text{Cl}_2].4\text{H}_2\text{O}$	6.9-7.6(m)[7H]	7.0-7.8(m) [14H]	6.6-7.8(m) [7H]	6.8-7.5(m) [14H]
$\text{Cu}L_2\text{Cl}_2].4\text{H}_2\text{O}$	6.9-7.6(m)[7H]	7.0-7.8(m) [14H]	6.6-7.8(m) [7H]	6.8-7.5(m) [14H]

**Legend:** m = medium; s = sharp

### $^{13}\text{C}$ -NMR

The  $^{13}\text{C}$ -NMR data of  $L_1$  and  $L_2$  and the synthesized metal complexes were recorded in DMSO. The phenolic carbon shifted to a lower field region in complexes proving co-ordination of the C-O group to the transition metal to form C-O-M bond. There was no significant shift due to the C atoms on bonding to the metal.

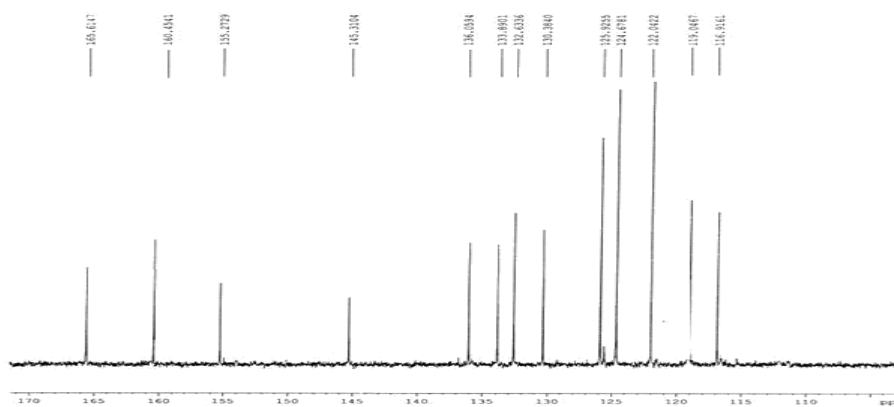
The C-7 azomethine carbon showed a down field shift of electron density from N to the metal to form N-M bond.

**Table 4:**  $^{13}\text{C}$ -NMR spectral studies of  $\text{L}_1$ ,  $\text{L}_2$  and its metal complexes

C/No	(1)	(5)	(7)	(11)
$\text{L}_1$	148.6.	148.5	148.9	149.7
$\text{L}_2$	145.0	145.0	145.0	146.1
$[\text{CuL}_1\text{Cl}_2].4\text{H}_2\text{O}$	118.4	118.0	118.2	119.4
$[\text{CuL}_2\text{Cl}_2].4\text{H}_2\text{O}$	123.4	123.0	121.2	121.4
$[\text{ZnL}_1\text{Cl}_2].4\text{H}_2\text{O}$	131.3	131.4	130.5	130.3
$[\text{ZnL}_2\text{Cl}_2].4\text{H}_2\text{O}$	130.1	130.1	119.4	119.0

**Table 5:** FTIR Spectra for  $\text{L}_1$ ,  $\text{L}_2$  and its metal complexes.

Codes	O – H	C = N	M – O	M – N	M – Cl
$\text{L}_1$	3473	1960	-	-	-
$\text{L}_2$	3477	1997	-	-	-
Cu	$[\text{CuL}_1\text{Cl}_2].4\text{H}_2\text{O}$	3436	1621	506	670
	$[\text{CuL}_2\text{Cl}_2].4\text{H}_2\text{O}$	3436	1576	332	449
Zn	$[\text{ZnL}_1\text{Cl}_2].4\text{H}_2\text{O}$	3473	1565	477	871
	$[\text{ZnL}_2\text{Cl}_2].4\text{H}_2\text{O}$	3481	1628	685	966

**Figure 2a:**  $^{13}\text{C}$  NMR spectrum of Schiff base formed with  $\text{L}_1$

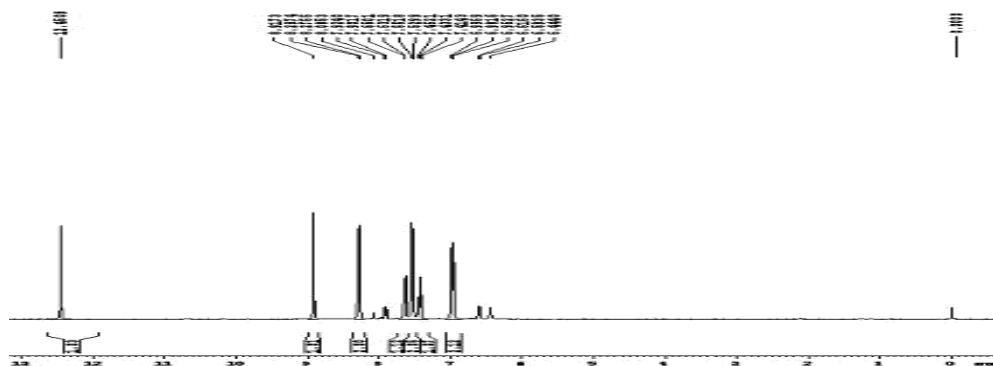


Figure 2b:  $^{13}\text{C}$ NMR of Schiff base formed with  $L_2$

### Fourier Transform Infrared Resonance (FTIR)

Using KBr pellets in the range  $4000 - 450 \text{ cm}^{-1}$ , the FTIR spectra were recorded and the characteristic bands are listed in the Table 5. Allocation of various vibration bonds were made by comparison of ligands spectra with that of the metal ions. The  $-\text{OH}$  vibrational stretch for the ligands occurred at  $3473$  and  $3477 \text{ cm}^{-1}$ . After complexation, the signals shifted downward in all the metal complexes. Due to the paramagnetic effect of all the  $\text{M}(\text{II})$  ions, there was a downward shift of the signals and hence support the coordination of the Schiff bases to the metal ions. At the lower region, the bonds allocated to the  $\text{M} - \text{N}$  bond ranges from  $400$  to  $477 \text{ cm}^{-1}$  and for  $\text{M} - \text{Cl}$  the bond allocated ranges from  $332$  to  $440 \text{ cm}^{-1}$ .

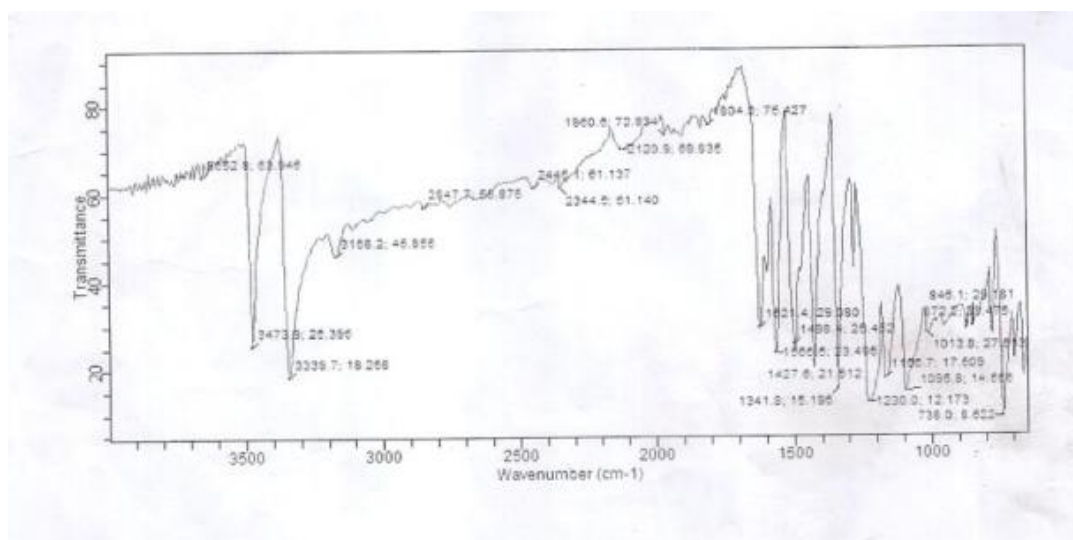


Figure 3a: FTIR Spectrum for 2 – nitroaniline

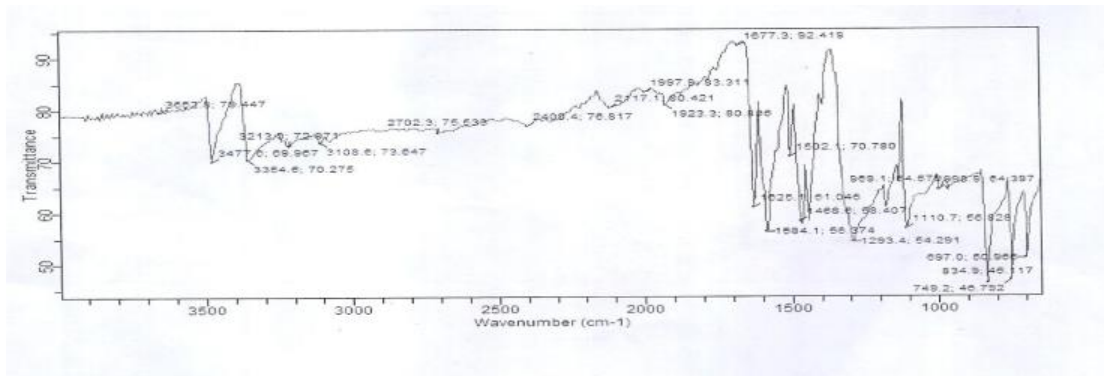


Figure 3b: FTIR spectrum for 4 – nitroline

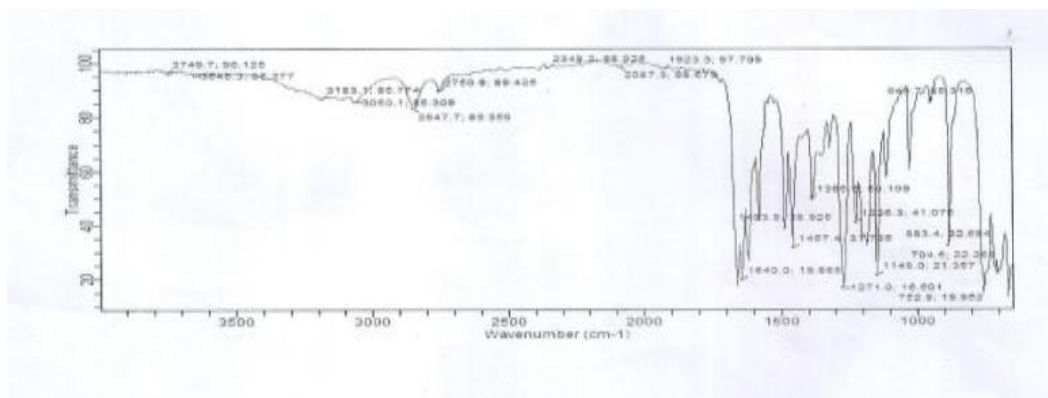


Figure 4a: FTIR Spectrum for Schiff base L<sub>1</sub>

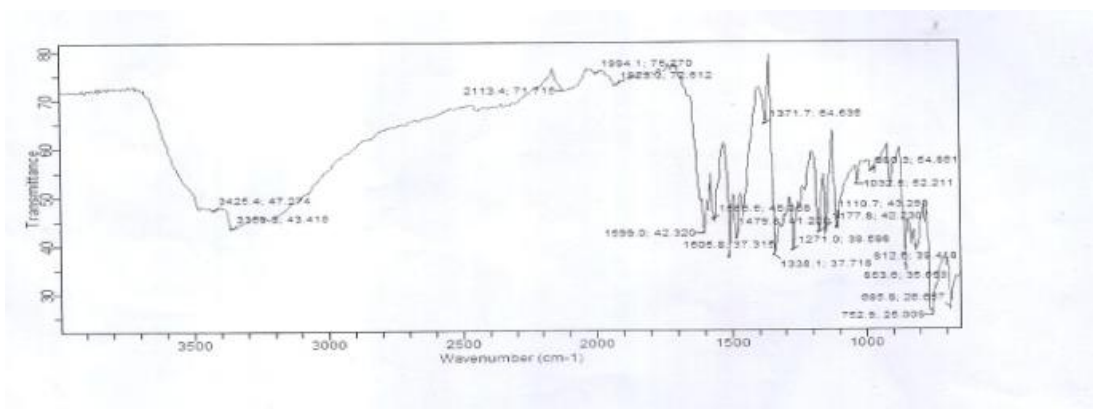


Figure 4b: FTIR Spectrum for Schiff base L<sub>2</sub>

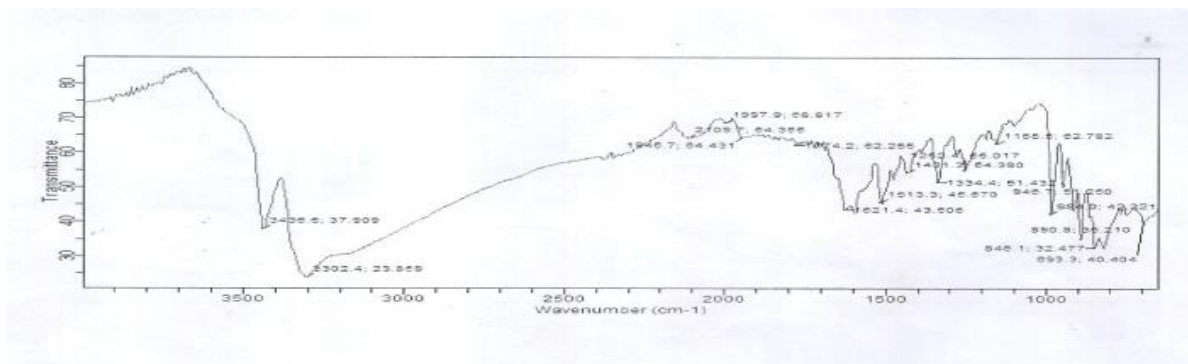


Figure 5a: FTIR Spectrum of the complex  $[CuL_1Cl_2].4H_2O$

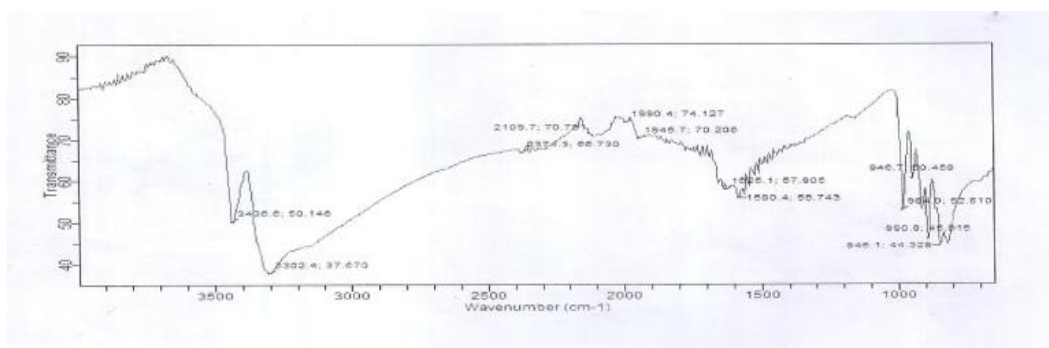


Figure 5b: FTIR Spectrum of the complex  $[CuL_2Cl_2].4H_2O$

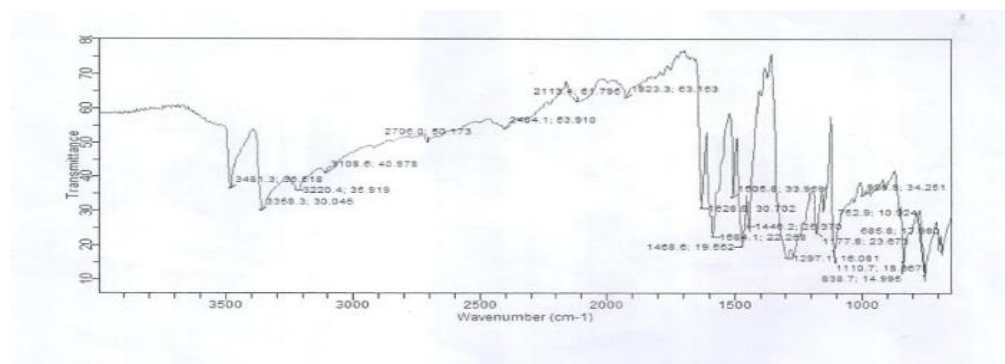
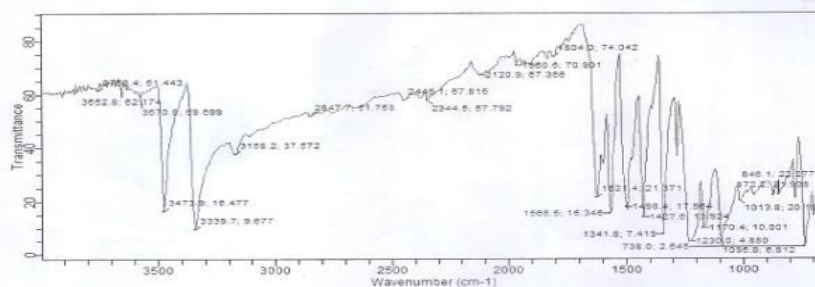


Figure 6a: FTIR Spectrum of the complex  $[ZnL_1Cl_2].4H_2O$



**Figure 6b:** FTIR Spectrum of the complex  $[ZnL_2Cl_2].4H_2O$

### Evaluation of nematocidal studies of *M. aranaria* using the synthesized ligands and its metal complexes

The analyses were carried out at five different concentrations ranging from 125 - 7.8ppm for one week. For  $L_1$ , the corrected percentage mortality of 16 percent was observed at a concentration of 7.8 ppm, but at higher concentration of 125 ppm and 62.5 ppm, the corrected percentage mortality was 83.0 and 80.7, respectively. On day 5, the corrected percentage mortality of 85.0 and 80.0 were observed at concentration of 125 ppm and 625 ppm, respectively. At lower concentrations of 15.3 ppm and 7.8 ppm, the corrected percentage mortality obtained was 35.3 and 17.9, respectively. At lower concentrations of 15.3 and 7.8 ppm, the corrected percentage mortality became 35.3 and 17.9, respectively. On the seventh day of treatment at higher concentration of 125 and 62.5ppm, the corrected percentage mortality recorded 89.0 and 80.3, respectively, while at lower concentration of 15.3 and 7.8ppm the corrected percentage mortality dropped to 37.3 and 19.3, respectively. A similar trend was observed for the Schiff base  $L_2$ , at lower concentrations of 7.8 and 15.3 ppm, the corrected percentage mortality taken was 35.0 and 30.00, respectively, while at a relatively higher concentrations of 125 and 62.5 ppm, the corrected percentage mortality moved to 87.2 and 86.2, respectively. On the seventh day, the percentage mortality was 90.2, 89.0, 63.3, 39.00 and 23.6 at the respective concentrations of 125, 62.5, 31.25, 15.25 and 7.81ppm. These were in resonance with what was obtained elsewhere [16], [26]. On the first day of treatment, the complex  $[CuL_1Cl_2].4H_2O$  at concentration of 125.0 and 62.5 ppm had a corrected percentage mortality of 99.7 and 99.3 and at concentrations of 15.3 and 7.8 ppm, the corrected percentage mortality were 60.0 and 50.0. These observations show that an increase in concentration increases the mortality of the nematodes. The complex  $[CuL_2Cl_2].4H_2O$  at



concentrations of 15.3 and 7.8 ppm on the first day of treatment, the corrected percentage mortality was 50.3 and 34.4 respectively. The corrected percentage mortality was 95.7 and 94.0 at 125 and 62.5 ppm, respectively. On the fifth day of treatment the corrected percentage mortality for  $[\text{CuL}_2\text{Cl}_2].4\text{H}_2\text{O}$  was 61.3 and 44.3 at 15.3 and 7.8 ppm respectively. On the seventh day of treatment, the corrected percentage mortality moved to 99.7, 97.7, 92.3, 70.0 and 45.3 respectively. These trends agreed with the findings obtained by Ugama [26]. The metal complex  $[\text{ZnL}_1\text{Cl}_2].4\text{H}_2\text{O}$  had 44.0 and 43.9 corrected percentage mortality at 15.3 and 7.8 ppm on the first day of treatment, whereas at concentrations of 125 and 62.5 ppm, the corrected percentage mortality were 95.6 and 95.0, respectively. On the third day of treatment, the corrected percentage mortality at 15.3 and 7.8 ppm was 57.0 and 40, respectively whereas at concentration of 125 and 62.5 ppm, the corrected percentage mortality was 97.0 and 91.0 respectively. At concentrations of 125 and 62.5 ppm the corrected percentage mortality recorded 98.0 and 94.05 respectively on the fifth day of treatment whereas at 15.3 and 7.8 ppm the corrected percentage mortality was 53.0 and 38.0, respectively. For metal complex  $[\text{ZnL}_2\text{Cl}_2].4\text{H}_2\text{O}$ , the corrected percentage mortality at 125 and 62.5 ppm was 98.3 and 80.0 on the first day of treatment whereas at concentrations of 15.3 and 7.8 ppm the corrected percentage mortality were 49.3 and 40, respectively. On the third day of treatment, the corrected percentage mortality became 98.6 and 91.0 at concentrations of 125.0 and 62.5 ppm, respectively, whereas at concentrations of 15.3 and 7.8 ppm, the corrected percentage mortality was 37.0 and 30.0, respectively. At the concentrations of 125 and 62.5 ppm, the corrected percentage mortality reported was 99.0 and 95.0, respectively, on the fifth day whereas at concentrations of 15.3 and 7.8 ppm, the percentage mortality showed 53.0 and 32.3, respectively [26].

From the results of the various analyses, there was a gradual increase in corrected percentage mortality with increasing length of treatment and concentration. The results indicated that the synthesized metal complexes show higher bioefficacy on *M. arenaria* at higher concentration than the ligands  $\text{L}_1$  and  $\text{L}_2$  at the same level of concentration. Conversely, the Schiff base ligands at lower concentrations of 15.3 and 7.8 ppm the mortality rate was lower in the range of 30.0 to 35.0 respectively (see Table 6). The result was in complete agreement with the findings by Ugama [26].

**Table 6:** Nematocidal studies of *M. arenaria* using the synthesized metal complexes and Schiff bases L<sub>1</sub> and L<sub>2</sub>

Synthesized compounds	No of days	Corrected percentage mortality at various concentration				
		125.0 ppm	62.5 ppm	31.3 ppm	15.3 ppm	7.8ppm
L <sub>1</sub>	1	83.0	80.7	44.7	22.7	16.0
	3	84.0	81.0	61.3	32.7	16.3
	5	85.0	80.0	62.0	35.3	17.7
	7	89.0	80.3	66.3	37.3	19.3
Control	1	0	0	0	0	0
	3	0	0	0	0	0
	5	0	0	0	0	0
	7	0	0	0	0	0
L <sub>2</sub>	1	87.2	86.2	36.3	35.0	30.0
	3	87.5	87.0	55.7	28.3	22.0
	5	88.6	88.0	57.3	34.7	25.3
	7	90.2	89.0	63.3	39.0	23.6
[CuL <sub>1</sub> Cl <sub>2</sub> ].4H <sub>2</sub> O	1	99.7	91.3	64.7	60.0	50.0
	3	99.7	94.7	74	72	25.7
	5	99.7	97.0	77.7	46	44.0
	7	100.0	92.0	78	47	35
[CuL <sub>2</sub> Cl <sub>2</sub> ].4H <sub>2</sub> O	1	95.7	94.0	64.7	50.3	34.5
	3	97.3	96.3	75.0	56.3	40.3
	5	99.3	96.3	92.7	61.3	44.3
	7	99.7	97.7	92.3	70.0	45.3

[ZnL <sub>1</sub> Cl <sub>2</sub> ].4H <sub>2</sub> O	1	95.6	95.0	49	40.0	38.0
	3	97.0	91.3	51.0	57.0	40.0
	5	98.0	94.5	73.0	53.0	38.0
	7	100.0	99.4	91.0	62.0	42.0
[ZnL <sub>2</sub> Cl <sub>2</sub> ].4H <sub>2</sub> O	1	98.3	80.0	52.0	49.3	40
	3	98.6	91.0	52.3	37.3	30.0
	5	99.0	95.0	73.3	53.0	32.3
	7	100.0	96.0	91.0	62.3	42.3

The results showed the efficiency of the synthesized ligands and their metal complexes which is similar to studies undertaken by elsewhere [10][11] who reported that metal complexes had high bioefficacy against root knot nematodes. The application of the ligands and their metal complexes can suppress the population of *M. arenaria* and promote the growth and yield of *Arachis hypogea*. This is attributable to the high nutrient content that will become available to the peanuts. This result is consistent with researches previously carried out by elsewhere,[12], [13] [26].

## Conclusion

The research has achieved the synthesis, characterization and nematocidal activities of metal complexes and some aldimine Schiff bases against *M. arenaria*. In this study, the root knot nematode, *M. arenaria* was identified and confirmed to dominate pests affecting *Arachis hypogea*. The synthesized aldimine ligands and their metal complexes effectively inhibited the activities of the nematodes and sacrificing them in the process. In comparison, the metal complexes showed higher inhibiting, sacrificing ability and potency than the free ligands in suppressing the repopulation of nematodes.

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