

# Synthesis, characterization and biological profiles of Schiff base aldimines derived from N-N<sup>1</sup>-Diphenyl-O-Pyrol-6-Methyleneacetate and its Cu<sup>2+</sup> and Fe<sup>2+</sup> complexes

Iorungwa, M. S.\*, Atagher, J. A., Shimaibo, P. T. and Nanev, J. D.

Inorganic/Physical Chemistry Research Group, Department of Chemistry, Federal University of Agriculture, Makurdi – 970001, Benue State Nigeria.

\*Corresponding author. Email: [lorungwa.moses@uam.edu.ng](mailto:lorungwa.moses@uam.edu.ng); Tel: +2347039189185.

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**ABSTRACT:** The aldimine ligand N-N<sup>1</sup>-diphenyl-o-pyrol-6-methyleneacetate (PPA) was prepared by a condensation reaction between 2-phenylglycinemethylesterhydrochloride and pyrol-2-carbaldehyde which was used to synthesize transition metal complexes of Cu(II) and Fe(II) employing the microwave assisted technique. The ligand and complexes were characterized by FTIR, UV, XRD, and magnetic susceptibility measurements. Spectral results which had PPA – Cu (46.619 cm<sup>-1</sup>) and PPA – Fe (43.859 cm<sup>-1</sup>) revealed complexation arising from the nitrogen of the azomethine group. The UV and magnetic susceptibility confirmed the probable geometries of Cu(II) and Fe(II) complexes to be octahedral with magnetic moments of 1.66 and 1.11 B.M respectively. Crystallographic diffraction pattern revealed the unit cell dimension of a (7.9Å), b(7.4Å), c(7.0Å), α(90°), β(90°), γ(90°) for Fe and a(4.3 Å), b(3.8Å), c(4.46Å), α(90°), β(90°), γ(90°) for Cu(II) aligning with orthorhombic crystal system. Molar conductance measurement of 717 and 229 Ω<sup>-1</sup>cm<sup>2</sup>mol<sup>-1</sup> PPA – Cu and PPA – Fe respectively indicated that the complexes are electrolytic in nature. The metal complexes showed higher inhibiting potential compared to the free ligand when tested against the root knot nematode *Meloidogyne incognita* present in the crop *Esulentum spp* implying that the complexes can be used as nematicides.

**Keywords:** 2-phenylglycinemethylesterhydrochloride, *Esulentum spp.*, Schiff base, *Meloidogyne incognita*, pyrol-2-carbaldehyde.

## INTRODUCTION

The synthesis and study of metal complexes containing biologically relevant ligands with azomethine linkage have received great attention, and is encouraged by the importance of metal ions in a variety of biological processes. Besides their wide range of applications in food industry, these classes of compounds have demonstrated substantial catalytic and analytical behaviours and have also being harnessed in the area of research for new drug design (Iorungwa *et al.*, 2020).

The formation of complex compound is through a donor-acceptor interaction involving a central metal atom/ion with vacant orbitals and a molecule or an ion possessing a lone pair of electrons. The bond resulting from this type of

interaction is called dative covalent or coordinate covalent bond, and the compound thus formed is known as coordination compound or simply a complex (Akter *et al.*, 2017). The ability of transition elements to form complex compounds is attributed to the presence of vacant (n-1)d orbitals and existence of variable oxidation states. The formation of azomethine ligands most commonly referred to as Schiff base proceeds in a sequence of two types of reactions, including the nucleophilic addition of the amine group to the aldehyde or ketone to give carbinolamine as unstable addition product (Ruanpanun *et al.*, 2011). Amongst other factors, the stability of complexes is dependent on the nature of the ligand (Da Silva *et al.*,

2011). Schiff bases are a class of polydentate ligands capable of forming very stable complexes with transition metals. Their chelating ability combined with the ease of preparation and flexibility in varying the chemical environment; coupled with their moderate electron donation and easy tunable electronic and steric effects also make Schiff bases as versatile ligands capable of stabilizing different metals in various oxidation states (Da Silva *et al.*, 2011). Recently, some metal complexes containing anilidoaldimine ligands have been reported and have been shown to demonstrate catalytic activities in ring opening polymerization of cyclic esters (Chairany *et al.*, 2018). Many Schiff base complexes of metal ions are useful in various catalytic reactions such as oxidation, hydroxylation, aldol condensation and epoxidation.

The rate determining step which is the dehydration of carbinolamine is catalyzed by an acid of mid pH as amines are basic compounds (Atagher, 2021; Iorungwa *et al.*, 2021a). The azomethine nitrogen in the Schiff base not only provides the binding site for metal ions, but also enhances the attachment with various substrates of biomolecules like proteins, and amino acids in biological systems and that of disease causing agents (Bal *et al.*, 2014). Research findings in the area of interaction of metal ions with organic ligands have shown that the complex possess better antimicrobial activity compared to the free ligands achieved through chelation (Mahapatra *et al.*, 2014; Fei-Ran *et al.*, 2014; Deshpande and Seema, 2016). Schiff base thiodiazole derivatives with salicyl-aldehyde or o-vanillin and their metal complexes with Mo (II) shows insecticidal activities against boll worm and promote cell survival rate of mung bean sprouts (Anant and Devjani, 2011). Nematicidal activities of Schiff base metal complexes against different species of nematodes have also been demonstrated (Ekta *et al.*, 2014) still, their nematicidal potential against root knot nematode *M. incognita* needs to be explored further. The azomethine nitrogen in Schiff base, not only provides binding site for metal ions, but also make attachment with various substrate of bio-molecules like protein and amino acids in biological systems and that of disease causing germs (Narendra and Parashuam, 2013). Through the metabolic activities, our body generates Schiff base complexes that show activities against various microbes. The first-row transition metal (II) ion complexes of chromenyl Schiff base are associated with lots of biological activities such as anticancer and antifungal activities.

The discovery of new metal-based drugs has been largely based on the ability of metals to increase inhibitory potential of chemotherapy agents. Efficiency of some therapeutic agents has been reported to have increased upon coordination to transition metals (Chandraleka *et al.*, 2011). Transition metal complexes derived from drugs have been among the most widely studied coordination compounds in recent years, since they are becoming increasingly important as biochemical and antibacterial

and antifungal agents was reported by Goreci *et al.* (2016).

Bansal and Dave (2013) synthesized the mixed ligand complexes of  $Mn^{2+}$  and  $Zn^{2+}$  with phthalic acid or succinic acid and heterocyclic amines. The antibacterial activities of the complexes were investigated against the Gram-positive bacteria, *Bacillus cereus* and *Staphylococcus aureus*, and Gram negative *Escherichia coli* and *Shigella sonnei*. Ciprofloxacin was used as the standard antibacterial agent. The tested mixed-ligand complexes showed good bacterial effect. The structure of the synthesized complexes was determined by elementary analysis, magnetic moment measurements, conductometric, FTIR spectral studies and electronic spectral analysis. The  $Mn^{2+}$  complexes were paramagnetic with an octahedral geometry, and the  $Zn^{2+}$  diamagnetic with both tetrahedral and octahedral structures.

The microbial activity of the N-(2-hydroxy-1-naphthalidene) phenylglycine and its transition metal complexes was investigated. The antifungal screening data indicated that the activity of the ligand has increased upon complexation.  $Cu^{2+}$ ,  $Ni^{2+}$  and  $Co^{2+}$  complexes have demonstrated better antifungal activity compared to the ligands and corresponding metal salts.

This research is targeted at the synthesis, characterization and nematicidal studies of  $Cu^{2+}$  and  $Fe^{2+}$  complexes from 2-phenylglycinemethyl ester hydrochloride and pyrrol-2-carbaldehyde.

## MATERIALS AND METHODS

All the chemical and solvents used for the synthesis were of analytical grades and purchased from BDH and Merck Chemical Company Limited. They were used without further purification. The infrared spectra of the ligand and metal complexes were run on KBr disc in the range 4000-450  $cm^{-1}$  on a Shimadzu infrared spectrophotometer. The electronic transition was monitored using Perkin Elmer spectrophotometer UV-VIS double beam PC scanning spectrophotometer. Gallenkamp melting point apparatus with digital thermometer, conductivity meter was all used for characterization.

X-ray power diffraction characterization of the synthesized Schiff bases and their metal complexes were carried out at the Umaru Musa Yar'adua University Katsina State Nigeria in the Central Research Laboratory with X-ray Diffractometer Thermo Scientific Model ARL X TRA X-ray 197492086. The analysis was carried out to determine the type of crystal system lattice parameters and the cell volume.

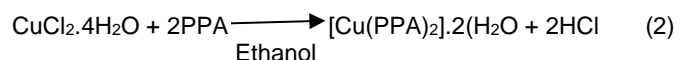
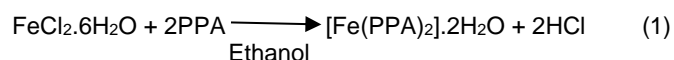
### Preparation of N-N<sup>1</sup>-diphenylpyrrol-6-methyleneacetate

The synthesis of N-N<sup>1</sup>-diphenylpyrrol-6-methyleneacetate was carried out in accordance with the method described

by lorungwa *et al.* (2020). Exactly 3.3 g (0.01 mmol) of 2-phenylglycinemethylester hydrochloride accurately weighed into a crucible and pulverized into fine powder, and 2.139 g (0.01 mmol) of pyrrol-2-carbaldehyde and 10 mL of absolute ethanol added. A 2.78 g (0.01 mmol) of triethylamine was added and the mixture, stirred in a beaker for 1 h and placed in the thermostated microwave oven DHG-9010-ISA PEC Medical USA model and power source of 220V for 30 min with continued stirring. The temperature of the oven was kept at 100°C and monitored using a thermocouple device. A white coloured solid product formed was filtered, washed with ethanol for several times, and oven-dried for 30 min. The solid crystals formed were dried in an oven at 50°C for 3 days and preserved in the refrigerator.

### Synthesis of complexes

Using 250 mL beaker, 5.0 g (0.01 mol) of Schiff base ligand and 20 mL ethanol was added drop wise into 1.0 mmol of metal salts of Cu(II) chloride and Fe(II) chloride respectively. Each mixture was stirred thoroughly and placed into a microwave oven for 30 min. The metal complexes formed were filtered and washed with distilled water. The products formed was air-dried and preserved in a refrigerator at 50°C (lorungwa *et al.*, 2020). The reaction between the Schiff base and the metal chlorides are presented in Equations 1 and 2 below:



### Nematicidal activity

The method for the nematicidal activities of the ligands and complexes as well as the thermogravimetric determinations was done as described in the previous research (lorungwa *et al.*, 2020) with slight modifications. Adult of *Meloidogyne incognita* suspect were collected from heavily infected unprotected farm lands in Mbaigba Council Ward in Tarka Local Government Area of Benue State and were conveyed in polyethylene bags to the microbiology laboratory of Federal University of Agriculture Makurdi for identification.

The nematodes were kept in properly labelled plates with 5 mL of distilled water. The Schiff base ligands and their metal complexes were prepared in the research laboratory of Chemistry Department.

The compounds were soaked in distilled water at different concentrations of 10.0, 5.0 and 2.5 ppm for 7 days. After filtration, 10 mL of the extract with various concentrations

were transferred to the Petri dishes containing 10 nematodes each. Equal volumes of distilled water were used as control.

The nematodes were exposed to the synthesized ligands and their metal complexes for 5, 10, and 15 h to study their nematicidal effects. The nematodes were considered dead if they did not move and were transferred to distilled water for 15 h to check if the nematodes remain dead. The corrected percentage mortality was calculated according to the equation:

$$\% \text{ Mortality} = \frac{\text{Mortality of treatment} - \text{Mortality of Co}}{1 - \text{Mortality of Co}} \times 100 \quad (3)$$

Where Co = zero concentration of ligands and complexes

The synthesized ligand PPA and its metal complexes of Cu and Fe were tested for mortality against the root knot nematodes (*Meloidogyne Incognita*) at three different concentrations which include 2.5, 5.0 and 10 ppm. This analysis was carried out within 25 h and observations were recorded every 5 h of exposure. In each of the treatment, exactly 10 juveniles were placed and their mortality rate observed, and their corrected percentage mortality rate calculated. For the complex PPA-Cu, it was observed that the corrected percentage mortality was 100% in all the concentrations of 2.5, 5.0, and 10 ppm and at various times of exposure.

## RESULTS AND DISCUSSION

The synthesized compounds were crystalline, coloured and soluble in water, acetone, DMF, methanol and ethanol. The spectroscopic data for the synthesized metal complexes were in agreement with the proposed geometry. Their physicochemical properties are as presented in Table 1 while the electronic spectral data and the corresponding unit cell parameters are captured in Tables 2 and 3 respectively

### UV/Visible studies

The electronic spectral studies of the ligands PPA, and its metal complexes were recorded in methanol. In the PPA spectra of the ligand, a sharp band appeared at 227 nm corresponding to n-π\* transition, with another band at 259 nm which underwent a blue shift to the shorter wavelength indicating complexation. The PPA-Fe complex showed a sharp band at lower energy and shifted hypsochromically to an absorption band of 273 nm.

The PPA-Cu complex showed absorption bands at 210 nm. Another broad band appeared at 220 nm which can be attributed to a π-π\* transition. Another absorption band appeared at 273 nm.

**Table 1.** Some physicochemical characteristics of ligands and complexes.

Ligand/Complex	Molecular Formula	Molecular weight	Colour	Yield(%)	pH
PPA	C <sub>15</sub> H <sub>14</sub> O <sub>2</sub> N <sub>2</sub>	254	White	88.2	6.4
PPA-Cu	[Cu(C <sub>15</sub> H <sub>14</sub> O <sub>2</sub> N <sub>2</sub> ) <sub>2</sub> ].2H <sub>2</sub> O	424.48	Black	11.8	2.4
PPA-Fe	[Fe(C <sub>15</sub> H <sub>14</sub> O <sub>2</sub> N <sub>2</sub> ) <sub>2</sub> ].2H <sub>2</sub> O	457.81	Brown	8.7	8.7

**Table 2.** Electronic spectral data for ligands/complexes.

Parameter	$\lambda_{\max}(\text{nm})$	$\lambda_{\max}(\text{cm}^{-1})$	Assignment	Geometry
PPA	227	44.052	n- $\pi^*$	-
	259	38.610	n- $\pi^*$	
PPA-Cu	210	46.619	$\pi - \pi^*$	Octahedral
	220	45.454		
	266	37.593		
PPA-Fe	228	43.859	$\pi - \pi^*$	Octahedral
	273	36.630		

**Table 3.** Unit cell parameters for synthesized ligands and complexes.

S/No	Ligand/Complexes	Unit Cell Parameters			Volume	Crystallite size	Crystal structure
		a(Å)	b(Å)	c(Å)			
1	PPA	4.2	4.1	4.9	93.63	24.27	Orthorhombic
2	PPA-Fe	7.9	7.4	7.0	409.22	47.96	Orthorhombic
3	PPA-Cu	4.3	3.86	4.46	74.03	1.76	Orthorhombic

The UV-visible absorption spectral studies of the ligands and their metal complexes are strongly correlated as an indication of similarities in their structures and geometry. The results are as presented in Table 2.

In the complexes, there were observable changes in both frequencies and intensities in the characteristic bands of the complexes compared to free ligands. The blue shifts in the absorption bands during complex formation indicate coordination of the ligands to the metal ion. These observations agreed with those of Gnanaprakash *et al.* (2016) and Jisha and Isaac (2017).

### FTIR characterization

The infrared spectral for the ligands and synthesized complexes were recorded using KBr pellets in the range of 4000-450  $\text{cm}^{-1}$  and provided relevant information with regard to the nature of the functional groups attached to the metal ion. The comparison of the IR spectral of the synthesized ligand and metal complexes showed the binding mode of the ligands to the metal ion which is confirmed by the shift in the positions of the absorption peaks.

The presence of absorption band above 3300  $\text{cm}^{-1}$  for the PPA-complexes indicates the presence of water molecules in those complexes as coordinated ligands. A band at 1494  $\text{cm}^{-1}$  is assigned to the spectrum  $\nu\text{C}=\text{O}$  stretching frequency in the spectrum of the free Schiff base ligand as seen in Figure 1 which is shifted to a lower frequency of this band indicates involvement of oxygen atoms from the COOH which bond to the metal ions. The peaks between 1576-1580  $\text{cm}^{-1}$  have been assigned to the  $\nu\text{C}=\text{N}$  mode (Hayat *et al.*, 2020). The shifting of this group to a lower frequency compared to the Schiff base ligand ( $\approx 1576 \text{ cm}^{-1}$ ) suggest a metal ion coordination through the nitrogen atom of the azomethine group. This band  $\text{C}=\text{N}$  (azomethine) shifted to a lower frequency due to the drift of lone pair density towards the metal centre as was reported by Hossein and Mohsen (2010).

The bands observed in the complexes around 1494 – 1490  $\text{cm}^{-1}$  region are assignable to  $\nu\text{C}=\text{O}$  (Iorungwa *et al.*, 2019). Other absorption bands appearing at 887 – 809  $\text{cm}^{-1}$  and 670  $\text{cm}^{-1}$  have been assigned to the M-N and M-O respectively. These results are in complete agreement with those of Yokeswari *et al.* (2019) and Gülcan *et al.* (2012).

The presence of  $\nu\text{C}=\text{N}$  and  $\nu\text{C}=\text{O}$  modes in the ligands

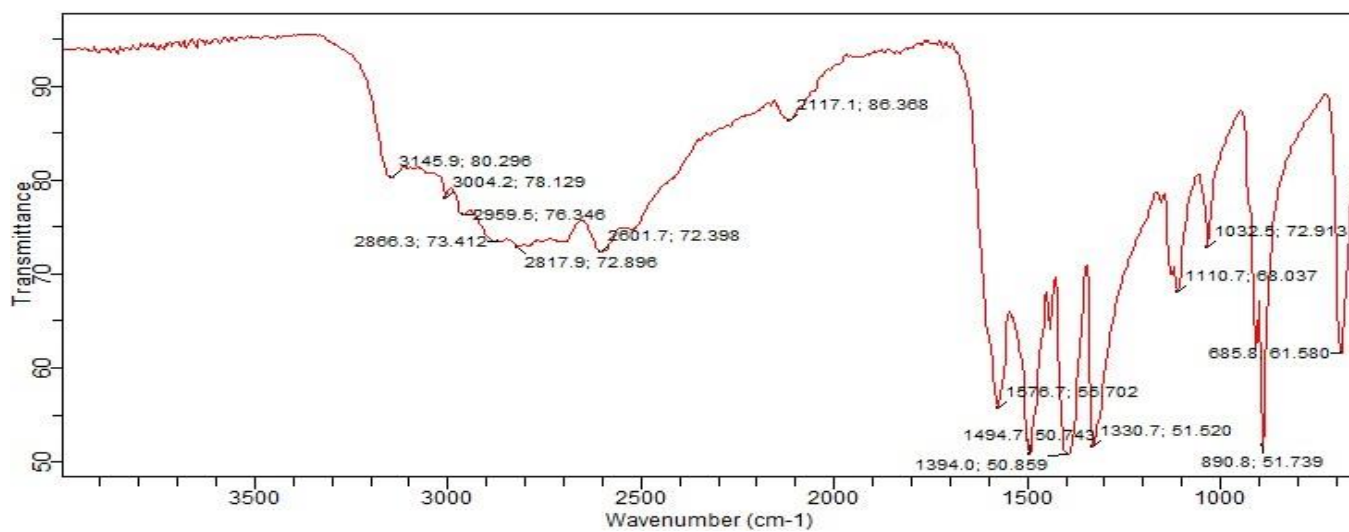


Figure 1. IR spectra of PPA ligand.

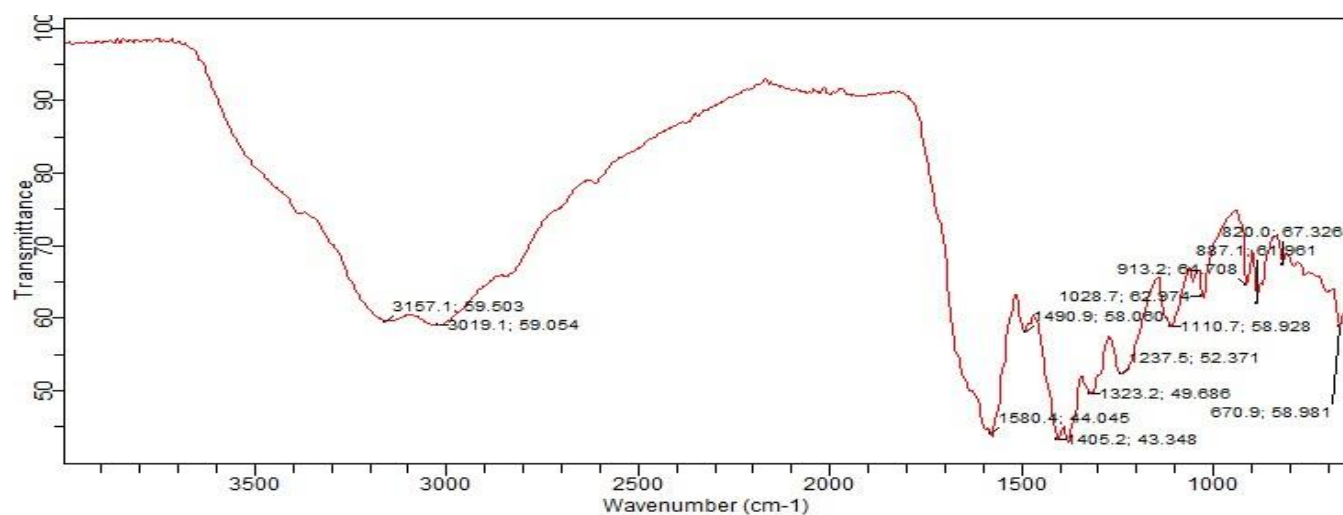


Figure 2. IR spectral of PPA-Cu complex.

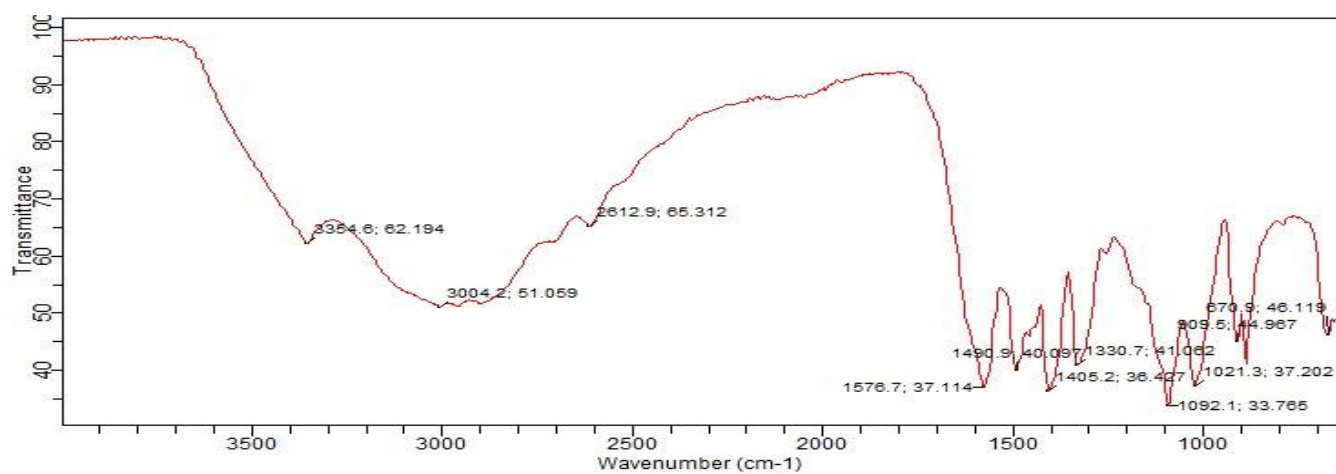


Figure 3. IR spectra of PPA-Fe complex.

**Table 4.** X-Ray diffraction studies of  $[\text{Fe}(\text{C}_{15}\text{H}_{14}\text{O}_2\text{N}_2)\text{Cl}_3]\cdot 4\text{H}_2\text{O}$  complex.

S/No	d-Spacing (Å)		2θ values		Δ2θ	hkl
	Observed	Calculated	Observed	Calculated		
1	5.03108	5.02479	8.8145	8.8008	0.0137	110
2	4.40355	4.39824	10.0829	10.0670	0.0159	111
3	3.96661	3.96172	11.2072	11.1921	0.0151	020
4	3.71597	3.71137	11.9739	11.9583	0.0156	200
5	3.51278	3.50861	12.6777	12.6561	0.0216	020
6	2.99079	2.98715	14.9378	14.9158	0.0220	211
7	1.97176	1.96941	23.0161	22.9856	0.0305	320
8	1.82942	1.82719	24.9237	24.8851	0.0386	030
9	1.62941	1.62742	28.2384	28.2032	0.0352	311
10	1.26296	1.26142	37.6201	37.5605	0.0596	300

**Table 5.** X-Ray diffraction studies of  $[\text{Cu}(\text{C}_{15}\text{H}_{14}\text{O}_2\text{N}_2)\text{Cl}_3]\cdot 2\text{H}_2\text{O}$  complex.

S/No	d-Spacing (Å)		2θ values		Δ2θ	hkl
	Observed	Calculated	Observed	Calculated		
1	4.35516	4.35003	10.1961	10.1834	0.0127	100
2	4.21302	4.20788	10.5439	10.5270	0.0169	001
3	3.86809	3.86332	11.4965	11.4784	0.0181	010
4	2.84492	2.84153	15.7231	15.7059	0.0172	110
5	2.74223	2.73894	16.3279	16.3019	0.0260	111
6	2.65440	2.65124	16.8842	16.8579	0.0263	111
7	2.23705	2.23434	20.1587	20.1329	0.0258	200
8	1.58362	1.58172	29.1317	29.0911	0.0406	220

and their shifts on complexation confirms (as appears in Figures 2 and 3) the ligands act as an N and O donor Schiff bases as collaborated by Gülcan *et al.* (2012).

### X-ray diffraction (XRD) characterization

As presented in Tables 4 and 5, the XRD pattern indicated a crystalline nature for the ligands and its metal complexes. Indexing of the diffraction pattern was performed by using the trial and error method. The Miller indices (*hkl*) along with observed and calculated 2θ angles, the observed and calculated *d* values are presented in Tables 4 and 5 and the spectral data (diffraction patterns) are presented in Figures 4, 5 and 6 for the free ligand and the complexes.

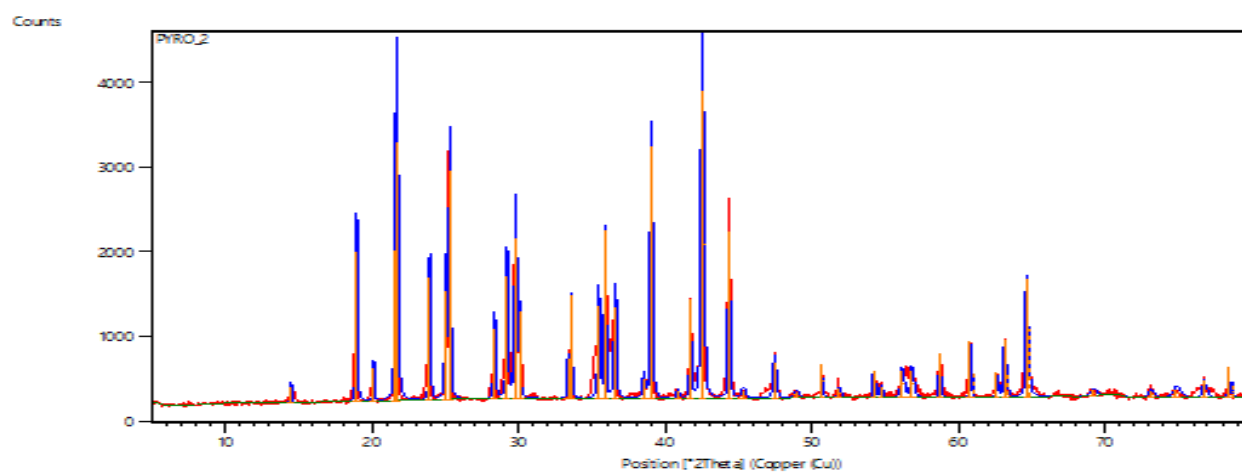
From the indexed data, (Tables 4 and 5), it was observed that the ligands and its metal complexes have features keeping with orthorhombic structure. The crystal structures of similar type of samples were reported as orthorhombic (Nwabueze and Salawu, 2012). From the diffraction data, the mean crystallite sizes of the ligands and complexes *D* was determined according to the Scherrer Equation as presented below:

$$D = \frac{0.9\lambda}{\beta \cos\theta} \quad (4)$$

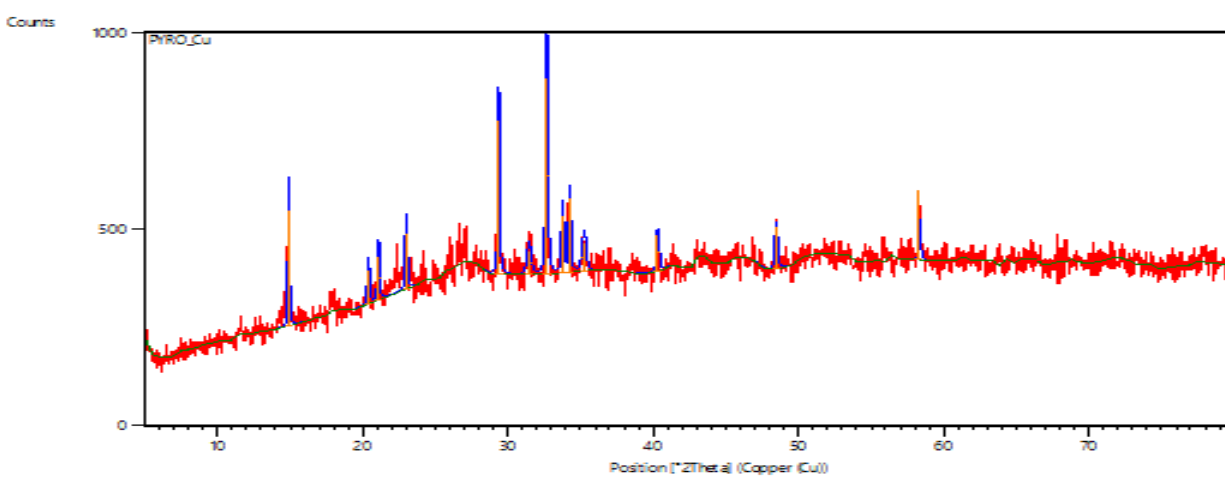
Where  $\lambda$  is the x-ray wavelength (1.5406 Å),  $\theta$  is the Bragg diffraction angle and  $\beta$  is the full width at half maximum of diffraction peak (Hayat *et al.*, 2020; Hossein and Mohsen, 2010). The average crystallites sizes of all the samples were found to be (48.28 nm) as presented in Tables 4 and 5.

### Thermo gravimetric Analysis

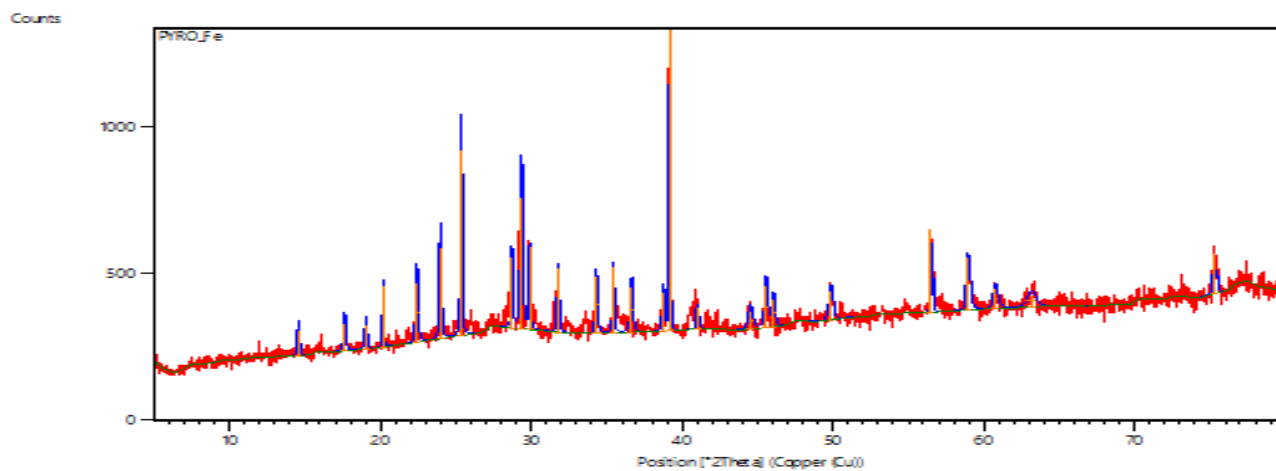
The thermal gravimetric analysis (TGA) and differential thermal analysis (TDA) experiments were carried out on the complex to determine the thermal stability of the complexes. The thermal behaviour of all the synthesized metal complexes was studied in the temperature range of 30-1000°C. Thermal analysis plays a vital role in studying the stability, melting point, structure and decomposition properties of the metal **complexes** (Iorungwa *et al.*, 2021b). The thermo gravimetric analysis provides information about thermal stability of the complexes and decides whether the water molecules are inside or outside the inner coordination sphere of the central metal ion. The



**Figure 4.** XRD measurement of PPA ligand.



**Figure 5.** XRD measurement of PPA-Cu complex.



**Figure 6.** XRD measurement of PPA-Fe complex.

**Table 6.** Thermoanalytical results for PPA complexes.

Name of complex	TGA (°C)	DTA (°C)	Stage	Mass loss		Assignment
				Found	Cal	
PPA-Cu [Cu(C <sub>15</sub> H <sub>14</sub> O <sub>2</sub> N <sub>2</sub> ) <sub>2</sub> ].2H <sub>2</sub> O	30-240	50	I	6.10	6.29	Dehydration of 2 moles of water
	241-470	450	II	84.6	86.01	C <sub>30</sub> H <sub>28</sub> O <sub>3</sub> N <sub>4</sub>
	>471	690	III	9.60	13.98	CuO Residue
PPA-Fe [Co(C <sub>15</sub> H <sub>14</sub> O <sub>2</sub> N <sub>2</sub> ) <sub>2</sub> ].2H <sub>2</sub> O	30-280	80	I	6.0	6.30	Dehydration of 2 moles of water
	281-450	380	II	66.0	87.23	C <sub>30</sub> H <sub>28</sub> O <sub>3</sub> N <sub>4</sub>
	>450	770	III	15.6	12.7	FeO Residue

results of the thermal behavior of the synthesized metal complexes are presented in Table 6. The result indicated a reasonable correlation between the calculated and found weight loss values.

The simultaneous TGA/DTA analysis of all metal complexes were studied where the heating rates were suitably controlled at 10 min under nitrogen atmosphere and weight loss was measured from ambient temperatures to 1000°C. The decomposition temperature range, percent mass loss and residue leaving behind are presented in Table 6. The thermogravimetric analysis of PPA-Fe with the formula [Fe(C<sub>15</sub>H<sub>14</sub>O<sub>2</sub>N<sub>2</sub>)<sub>2</sub>].2H<sub>2</sub>O indicated a weight loss of 6.0% found and 6.30% calculated associated with dehydration of two molecules of water at the temperature of 30-280°C. The second decomposition stage proceeded at 281-450°C corresponding to the loss of C<sub>30</sub>H<sub>28</sub>O<sub>3</sub>N<sub>4</sub>. Percentage weight loss of 66.0% found and 87.23% calculated strongly correlated. The third stage of decomposition corresponding to FeO residue at the temperature range of above 450°C, with weight losses of found 15.6% and calculated 12.7%.

The thermo gravimetric curve of the complex Cu(C<sub>15</sub>H<sub>14</sub>O<sub>2</sub>N<sub>2</sub>)<sub>2</sub>.2H<sub>2</sub>O shows a thermal decomposition at a temperature range of 30-240°C, weight loss of 6.10% found, 6.29% corresponding to the total mass loss of two molecules of water. Elimination of C<sub>30</sub>H<sub>28</sub>O<sub>3</sub>N<sub>4</sub> occurred at the second stage of the decomposition process at the temperature of 241-470°C with the weight loss of 84.6% found, 86.0% calculated. The third stage decomposition suffers the loss of ZnO residue at the temperature range above 471°C. In all cases, the residues are the metal oxides (Joshi *et al.*, 2011; Suman *et al.*, 2015). The percentage weight loss in all the complexes is in agreement with the calculated values (Kavitha *et al.*, 2015). The nature of the thermographs and percentage weight loss in all the complexes correspond to [ML<sub>2</sub>].3H<sub>2</sub>O, [ML<sub>2</sub>].2H<sub>2</sub>O, for PPA-Cu and PPA-Fe respectively. These observations are recorded elsewhere (Ekta *et al.*, 2014).

#### Evaluation of nematicidal studies of root knot nematodes (*Meloidogyne incognita*)

Table 6 shows the results of nematicidal studies of the root

knot nematodes of *Meloidogyne incognita* using Schiff base ligand and the synthesized complexes. The complex PPA-Fe showed that at 5 h of exposure, the corrected percentage mortality at a concentration of 2.5 ppm was 0%, and 33.3% at the concentration of 5.0 ppm, while the corrected percentage mortality of 77.8% was recorded at the concentration of 10 ppm. At the second five hours of exposure (10 h), the corrected percentage mortality at the concentration of 2.5 ppm was 11.1% whereas at the concentrations of 5.0 ppm and 10 ppm, the corrected percentage mortality were 44.4% and 99.7% respectively. At the last five hours of exposure (15 h), the corrected percentage mortality of 44.4% was recorded for the concentration of 2.5 ppm, but the higher concentrations of 5.0 ppm and 10 ppm, the corrected percentage mortality were 77.8% and 100% respectively. For the complex PPA-Cu, it was observed that the corrected percentage mortality was 100% in all the concentrations of 2.5, 5.0, and 10 ppm and at various times of exposure.

From the results of the analysis, the metal complexes showed higher efficiency compared to the free ligands in which the increase in concentration and time of exposure increases the mortality of the nematodes. This observation is in consonance with those recorded elsewhere (Ekta *et al.*, 2014; Karakaya *et al.*, 2016).

#### Conclusion

The synthesis of complexes of azomethine linkages have been extensively studied, and it is an area of research with great potentials in new drug design. The present work describes the synthesis of Cu<sup>2+</sup> and Fe<sup>2+</sup> Schiff base complexes derived from the condensation reaction of 2-phenylglycinemethylesterhydrochloride with pyrol-2-carbaldehyde. The physical (molar conductance, magnetic susceptibility measurement and thermal gravimetric analysis, spectral (uv-vis, FTIR) data for the complexes provides clear evidence that the Schiff base is coordinated to the metal ion through the azomethine linkage. It is generally observed that metal chelates exhibit higher inhibiting potentials for the tested nematodes compared to the free ligand as the chelates have increased cell permeability. It was also discovered that the efficacy of the



complexes were higher than those of the ligand at the higher concentrations and time of exposure.

## CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

## REFERENCES

- Akter, J., Hanifl, M. A., Saidul, M. I., Masuqul, M. H., Seung, H. L., & Laila, A. B. (2017). Synthesis, Characterization and antimicrobial activity of mixed ligand complexes of Mn (II) and Zn(II) with Phthalic Acid or Succinic Acid and Heterocyclic Amines. *Der Chemical Sinica*, 8(1),166-174.
- Anant, P., & Devjan, A. (2011). Application of Schiff bases and their complexes. A review. *International Journal of ChemTech Research*, 3(4), 1891-1896.
- Atagher, J. A. (2021). Synthesis, characterization and nematocidal studies of transition metal complexes and azomethines obtained from 2-phenylglycinemethylesterhydrochloride and some selected aldehydes. .Ph.D thesis submitted to the Department of Chemistry, Joseph Sarwuan Tarka University Makurdi. Pp. 1-217.
- Bal, M., Ceyhan, G., Avar, B., Kose, M., Kayraldiz, A., & Kurtoglu, M. (2014). Synthesis and X-ray diffraction, electrochemical and genotic properties of a new azo-Schiff base and its metal complexes. *Turkish Journal of Chemistry*, 38, 222-241.
- Bansal, N., & Dave, S. (2013). Analgesic and anti-inflammatory activities of Schiff base metal complexes- A review. *International Journal of Basic and Applied Chemical Science*, 3(1), 31-40.
- Chairany, S., Sudibyo M, & Abdul R (2018). Application of Fourier Transform Infrared (FTIR) spectroscopy coupled with multivariate calibration for quantitative analysis of curcuminoid in tablet dosage form. *Journal of Applied Pharmaceutical Science*, 8(8), 151-156.
- Chandraleka, G., Chandramohan, D., Dhanasekaran, D., & Panneerelvan, A. (2011). Antifungal activity of amino acid Schiff base copper (II) complexes with phenanthroline and bipyridyl. *International Journal of Chemical and Analytical Science*, 2(10), 1235-1240.
- Da Silva, C. M., da Silva, D. L., Modolo, L. V., Alves, R. B., de Resende, M. A., Martins, C. V., & de Fátima, Â. (2011). Schiff bases: A short review of their antimicrobial activities. *Journal of Advanced Research*, 2(1), 1-8.
- Deshpande, V. G., & Seema, I. H. (2016). X-ray diffraction study of transition metal complexes of heterocyclic Schiff bases. *Journal of Chemistry and Chemical Sciences*, 6(10), 934-944.
- Ekta, Utreja, D., & K Dhillon, N. (2014). Synthesis of metal complexes of schiff bases and their nematocidal activity against root knot nematode *Meloidogyne incognita*. *Letters in Organic Chemistry*, 11(2), 116-125.
- Fei-Ran, L., Xin.Z., Dong-Dong, W., Zhi-Ng. X., & Ying-Ke, M. (2014). Synthesis, spectral, thermal stability and bacterial activity of Schiff bases derived from alanine and threonine and their complexes. *Journal of Chemical and Pharmaceutical Research*, 7(8), 792-803.
- Gnanaprakash, S., Madhumtha, B., Jayapradha, C., Devipriya, S., & Kalaiarasan, P. (2016). Identification of resistance in mulberry, *Morus SPP*. For Root knot nematode, *Meloidogyne incognita*. *International Journal of Plant Science*, 2(2), 262-264.
- Goreci, C.Y., Zulal, D and Nilay A. (2016). Green synthesis of new amino acid Schiff base and their biological activities. *Journal of the Turkish Chemical Society*, 3(3), 15-26.
- Gülcan, M., Sönmez, M., & Berber, I. (2012). Synthesis, characterization, and antimicrobial activity of a new pyrimidine Schiff base and its Cu (II), Ni (II), Co (II), Pt (II), and Pd (II) complexes. *Turkish Journal of Chemistry*, 36(1), 189-200.
- Hayat, K. A., Yerramilli, S., Adrien, D., Terry L. A., Daria, C. B., & Gregory, S. P. (2020). Experimental methods in chemical engineering: X-ray diffraction spectroscopy—XRD. *Canadian Journal of Chemical Engineering*, 98,1255-1266.
- Hosseini, N., & Mohsen, M. (2010). Synthesis and characterization of nitro-Schiff bases derived from 5-nitrosalicylaldehyde and various diamines and their complexes of Co(II). *Journal of Coordination Chemistry*, 63(1), 156-162.
- lorungwa, M. S., Mamah, C. L., & Wuana, R. A. (2019). Synthesis, characterization, kinetic, thermodynamic and antimicrobial activity studies of complexes of Cd (II), Cr (III), and Zr (IV) derived from benzaldehyde and ethylenediamine. *ChemSearch Journal*, 10(2), 10-24.
- lorungwa, M. S., Ugama A. G., Wuana A. R., Amua, Q. M., Tivkaa, J., lorungwa, P. D., & Surma, N. (2021b). Nematocidal 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*. *International Journal of New Chemistry*, 8(3), 365-385.
- lorungwa, M. S., Wuana, A. R., Tyagher, L., Agbendeh, Z. M., lorungwa P. D, Surma, N., & Amua, Q. M. (2020). Synthesis, characterization, kinetics, thermodynamics and nematocidal studies of Sm(III), Gd(III) and Nd(III) Schiff base complexes. *ChemSearch Journal*, 11(2), 24-34.
- lorungwa, M. S., Wuana, R. A., Chahul, H. F., Atagher, J. A., & Ona, J. I. (2021a). Synthesis, characterization and nematocidal studies of Cu<sup>2+</sup> and Fe<sup>2+</sup> complexes of Schiff base derived from 2-phenylglycinemethylesterhydrochloride and benzaldehyde. *FUAM Journal of Pure and Applied Science*, 1(2), 99-109.
- Jisha, M. J., & Isaac, S. R. (2017). Synthesis and characterization of Schiff base complexes of Cu(II), Ni(II), Co(II) complexes of Schiff base derived from furan 3-carboxaldehyde and 3- amino pyridine. *International Journal of Scientific and Research Publications*, 7(10):10-19.
- Karakaya, C., Bede, B., & Cicek, E. (2016). Novel metal (II) complexes with bidentate Schiff base ligand: Synthesis, spectroscopic properties and dye decolorization functions. *Proceedings of the 50th Zakopane School of Physics, Zakopane, Poland*, 129(2),1208-2012.
- Mahapatra, B. B., Mishra, R. R., & Sarangi, A. K. (2016). Synthesis, characterisation, XRD, molecular modelling and potential antibacterial studies of Co (II), Ni (II), Cu (II), Zn (II), Cd (II) and Hg (II) complexes with bidentate azodye ligand. *Journal of Saudi Chemical Society*, 20(6), 635-643.
- Narendra, K. C., & Parashuram, M. (2013). Synthesis, characterization and molecular modelling of Ni(II) and cu(II) complexes with Schiff base derived from 1H-benzo[d]imidazole-4-amine and 2-hydroxy benzaldehyde. *Archives of Applied Science Research*, 5(5), 191-197.
- Nwabueze, J. N., & Salawu, O. W. (2012). Complexes of Ni(II), and Cu(II) sulphates with keto and enol forms of isobutryl

acetic acid, 4-amino benzoic acid and 4-cyno benzoic acid hydazide. *Advances in Pure and Applied Chemistry*, 1(2), 1-9.

Ruanpanun, P., Laatsch, H., Tangchitsomkid, N., & Lumyong, S. (2011). Nematicidal activity of fervenulin isolated from a nematicidal actinomycete, *Streptomyces* sp. CMU-MH021, on *Meloidogyne incognita*. *World Journal of Microbiology and Biotechnology*, 27, 1373-1380.

Yokeswari, N. P., Ananthi, P., & Shanmuga, P. D. (2019). Synthesis, Characterization and Biological activities of Schiff base Ligands and its Metal Complexes. *International Journal of Advanced Scientific Research and Management*, 4,77-81.