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Synthesis, Characterization and Antibacterial Activities of Mixed Ligand Metal(II) Complexes of Substituted Benzoic acid hydrazide and Isoniazid

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Abstract

Mixed ligand metal complexes of substituted benzoic acid hydrazide and isoniazid were synthesized and characterized by percentage metal analysis, magnetic susceptibility, UV-Visible, IR spectroscopy and conductivity measurements. The infrared study revealed that the isoniazid and the hydrazide derivatives coordinated via the amino $-NH_2$ group and the carbonyl oxygen =C-O, acting as bidentate ligands with a reaction stoichiometry of isoniazid: hydrazides derivatives: metal chloride salt in the ratio 1:1:1. The observed magnetic moment revealed the paramagnetic nature of the complexes of mixed isoniazid and benzoic acid hydrazide derivatives (NH_2 or OH) with values of 3.18 and 3.69 BM for Co and 2.41 and 3.05 BM for Ni and 1.78 and 2.16 BM for Cu respectively. The conductivity results indicated that the complexes are non-electrolytic in nature while the UV-visible displayed several transitions characteristic of complexes in the octahedral geometry. The ligands and complexes were screened for antibacterial activities against eight strains of pathogenic bacteria. The results showed that the ligand displayed higher antimicrobial activities than the complexes.

***Keywords*: Isoniazid, Antimicrobial, Metal(II) Complexes, Hydrazides**

Introduction

Hydrazides is a class of organic compounds with a common functional group characterized by nitrogen-nitrogen covalent bond with four substituents with at least one of them being an acyl/aryl group having a general formula NH_2-NH-R where R may be an acyl/aryl group. They constitute a versatile class of organic compounds with interesting biological properties such as anti-inflammatory (Al-Shaalan, 2011), anticonvulsant (Deepa and Aravindakshan, 2005), antituberculosis (Chohan and Kauser, 2000), antitumor (Aincough *et al.,* 1998), anti-HIV and antimicrobial activity and antiviral (Agwara *et al.,* 2010).

They are important compounds for drug design and as ligands for metal complexes, organo-catalysis and also important in the synthesis of heterocyclic compounds. These varying properties of carbonic acid hydrazides along with their applications in analytical chemistry have variously led to increased interest in their complexation characteristics with transition metal ions (Chohan *et al.,* 2006).

In coordination chemistry, hydrazides find their application as ligand forming chelates with metals, usually from the transition series. Studies have shown that the azomethine group having a lone pair of electrons in either a p or sp^2 hybridized orbital on trigonally hybridized nitrogen has considerable biological importance (Al-Shaalan 2007; Anwar *et al.,* 2011). Heterocyclic Schiff base ligands containing an additional azo functional group have varying applications in medicine (Lashanizadegan and Jamshidbeigi, 2012; Mitu *et al.,* 2012). They have been of great importance due to their synthetic flexibility, selectivity and sensitivity towards the metal ions. Those containing sulphur, nitrogen and oxygen impart special biological activity to these Schiff bases and their metal complexes (Singh and Varshney, 2006).

In this research work, mixed ligand metal complexes of substituted benzoic acid hydrazide and isoniazid were synthesized, characterized and their antibacterial activities against eight strains of pathogenic bacteria were investigated. These antibacterial activities were compared to that of the ligand, in order to ascertain the effect that coordination has on these activities.

Materials and Methods

Materials

The reagents and solvents: methyl-4-aminobenzoate, methyl-4-hydroxylbenzoate, hydrazine hydrate, ethylenediaminetetraacetic acid (disodium salt), DMSO, DMF, acetone, methanol, ethanol, ammonia and ammonium chloride, zinc sulphate, perchloric acid, hydrochloric acid, murexide dye, solochrome dye, copper(II) chloride, cobalt(II) chloride and nickel(II) chloride are of analytical grade and used as received.

Test Organisms

The microorganisms were maintained Muller-Hinton (MH) agar plates and stored at 4°C and later grown at 37°C when required. The strain used are of National Collection of Industrial Bacteria (NCIB) and Locally Isolated Organisms (LIO) from clinical samples. The locally isolated organisms were obtained from the Department of Microbiology, Obafemi Awolowo University Teaching Hospitals Complex, Ile-Ife, Nigeria. The organisms include: *Bacillus cereus* (NCIB 6349), *Escherichia coli* (NCIB 86), *Bacillus subtilis* (NCIB 3610), *Staphylococcus aureus* (NCIB 8588), *Salmonella typhi* (LIO), *Shigella spp* (LIO). The viability and purity of organisms were checked periodically by regular plating. Test cultures were prepared by aseptic transfer and grown at 37°C.

Culture Media

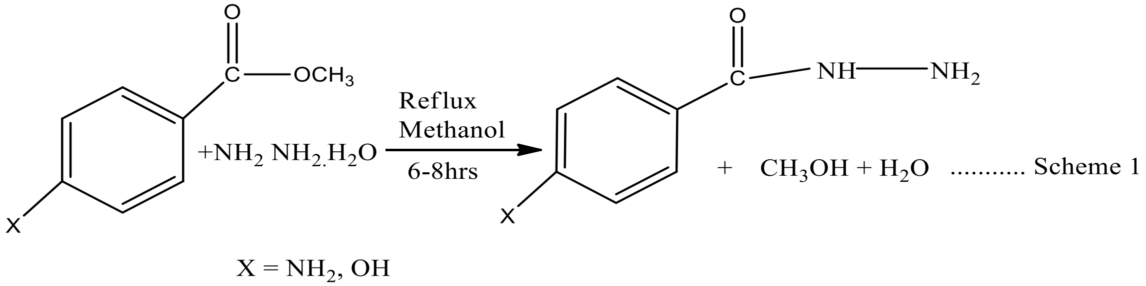
Rapid Labs Mueller Hinton Agar (CM-MHA 135) from United Kingdom, comprising per Litre: 17.5 g Peptone, 1.5 g Starch, 4.0 g Beef Infusion Solids and 15.0 g Agar. Himedia Nutrient Agar (M001-500G) from India, comprising per Litre: 5.0 g Peptone, 15.0 g Agar, 5.0 g NaCl, 1.5 g Meat Extract, 1.5 g Yeast Extract. Oxoid Nutrient Broth (CM-0001) from England, comprising per Litre: 5.0 g Peptone, 5.0 g NaCl, 1.0 g Lab-Lemco Powder. These culture media were purchased from Lyke Diagnostics England.

Physical Measurements

The infrared spectra were recorded on Shimadzu FT-IR 8000 Spectrometer range 400-4000 cm^{-1}. The electronic spectra of the complexes and the ligands were recorded in DMSO using Shimadzu UV-Vis Spectrometer. Melting points were determined on a Reichert Thermovar melting point apparatus; the conductivity measurement was done using Philips Pw. Digital Conductometer. The magnetic susceptibility measurement was carried out using Magnetic Susceptibility Balance (Sherwood Scientific Cambridge UK).

Synthesis of p-aminobenzoic acid hydrazide ligands (HL)

Methyl-4-aminobenzoate (10 g, 0.066 mol) in 50 ml ethanol was warmed for about 5 minutes until properly dissolved and added to Hydrazine hydrate (20 ml, 0.40 mol) dissolved in 50 ml of ethanol. The mixture was refluxed for about 8 hours and the resultant solution was left in a beaker for 2 days. White precipitates of p-aminobenzoic acid hydrazide (HL_1) was formed, collected and washed severally with methanol and then filtered to give white crystalline powder. The product was dried in a desiccator over anhydrous calcium chloride. Scheme 1 below represents the synthesis of the hydrazide derivatives. Methyl-4-hydroxybenzoate was employed for the synthesis of HL_2 using the same procedure.



Scheme 1: Synthesis of Substituted benzoic acid hydrazide

Synthesis of the mixed Metal(II) Complexes of HL

A mixture of p-aminobenzoic acid hydrazide – HL_1 (0.40 g, 0.003 mol) in 20 ml of methanol and isoniazid (0.72 g, 0.005 mol) in 20 ml of distilled water was stirred for 10 mins. A methanolic solution of copper(II) chloride dihydrate (0.45 g, 0.003 mol) was then added dropwise into the stirring homogenous mixture of the ligands and refluxed for 2 hours to produce a yellow solution which was allowed to stand for few hours. Precipitates of the metal complex was collected, washed with methanol and dried in a desiccator over anhydrous calcium chloride.

Antimicrobial Studies

This was carried out using the agar well diffusion method. Sterile MH agar plates were prepared and allowed to set and dry under laminar flow chamber. The bacterial population were prepared to correspond to 0.5 McFarland turbidity standard. The sterile MH plates were then innoculated with the standardized organisms using sterile cotton swabs. 6 mm sterile cork borer was used to bore wells into the agar, which were carefully filled with the solution of the synthesized complexes. The set-up was left to stand for 60 minutes before incubated at 37 °C for 24 hours. The diameter of the zones of inhibition were measured and recorded to the nearest millimeter. 10 mg/mL of the synthesized complexes was prepared in 10% dimethyl sulfoxide as dissolution solvent. Antibacterial activity of complexes was compared with 1 mg/mL Streptomycin as positive control, while 10% DMSO served as the negative control.

Results and Discussions

Physicochemical Parameters

Table 1 presents the physicochemical parameters of the synthesized compounds. The mixed ligand complexes displayed high melting points and a variety of colours ranging from black to light green. The theoretical % yield of the metal ions in the complexes showed a fairly good correlation with the experimental values. They were obtained at an appreciable yield ranging from 55 -72 %. Equation 1 showed the general reaction of isoniazid and two ligands with hydrated metal salts in methanol producing the mixed ligand metal complexes

$$MCl_2.XH_2O + HL + INH \xrightarrow{\substack{\text{Methanol}\\ \text{Reflux}}} [M(HL^1)(INH)Cl_2] + XH_2O \text{ ....1}$$

All the complexes synthesized were insoluble in water and other common solvents except dimethylsufoxide (DMSO). The molar conductivity measured in DMF are in the range 2.78 -8.56 $\Omega^{-1}cm^2mol^{-1}$, suggesting the non-electrolytic nature of these complexes (Geary, 1971).

Table 1: Physical Properties and Analytical Data for the Compounds

Compounds	Formula (Formula weight)	Colour	Melting Point. (^oC)	%Metal ion Found (Calculated)	% Yield	Λm ($\Omega^{-1}cm^2mol$
HL_1	$C_7H_9N_3O$	White	212-214	_	66	-
HL_2	$C_7H_8N_2O_2$	White	268-270	_	78	-
[Co(HL_1)Cl_2INH]	$C_{13}H_{16}N_6O_2CoCl_2$ (418.24)	Yellow	>300	12.16 (12.97)	70	5.40
[Ni(HL_1)(INH)]	$C_{13}H_{16}N_6O_2NiCl_2$ (418)	Black	>300	12.12 (11.76)	68	4.62
[Cu(HL_1)Cl_2(INH)]	$C_{13}H_{16}N_6O_2CuCl_2$ (422.85)	Yellow	260-262	16.41 (16.34)	72	8.56
[Co(HL_2)Cl_2(INH)]	$C_{13}H_{15}N_5O_3CoCl_2$ (419.27)	Light Green	>300	12.09 (12.48)	55	6.02
[Ni(HL_2)Cl_2(INH)]	$C_{13}H_{15}N_5O_3NiCl_2$ (419.03)	White	>300	11.30 (11.64)	69	2.78
[Cu(HL_2)Cl_2(INH)]	$C_{13}H_{15}N_5O_3CuCl_2$ (423.88)	Light Green	>300	16.21(16.34)	72	3.38

Infrared Spectra

The structurally significant IR bands for free ligand and its complexes are reported in Table 2 and comparison of the IR spectra of the ligands to those of the metal complexes provided evidence of coordination to the metal ion (Nakamoto,2009). In the infrared spectrum of HL_1, the band at 3500 cm^{-1} is assigned to ῡ(N-H) vibrational frequency, while the two doublets bands at 3400 and 3215 cm^{-1} are due to ῡ(NH_2) vibrational frequency which is characteristics of amino group in asymmetric and symmetric modes respectively. In the IR spectrum of HL_2, the bands 3422 and 3321 cm^{-1} are assigned to ῡ(OH) vibrational frequency respectively while the bands at 3200 cm^{-1} is attributed to ῡ(NH_2) vibrational frequency. The carbonyl stretching frequency band ῡ(C=O) at 1683 and 1635 cm^{-1} for both HL_1 and HL_2 respectively shifted to 1606 – 1746 and 1602 – 1614 cm^{-1}, while ῡ(NH_2) bands shifted to lower frequency in the metal complexes of HL_1 and HL_2 respectively suggesting coordination to the metal ion. In the infrared spectra for the synthesized hydrazide ligands, the bands at 1032 and 1025 cm^{-1} are attributed to ῡ(N-N) vibrational frequencies (Thaker *et al.*, 2005). Additional bands due to metal-oxygen and metal-nitrogen bonds that were absent in the spectrum of the ligands were observed in the spectrum of the metal complexes (Howlader *et al.*, 2009; Mostafa, 2011).

Table 2: Infrared Spectra Data (cm^{-1}) of the Hydrazides and their Metal Complexes

Compounds	ῡ(OH)	ῡ(NH)	ῡ(NH_2)	ῡ(C=O)	ῡ(C=N)	ῡ(N-N)	ῡ(M-O)	ῡ(M-N)
HL_1	_	3500w	3400s	1683m	1515m	1032w	_	_
HL_2	3422m	_	3200s	1635m	1514m	1025w	_	_
$[Co(HL_1)Cl_2(INH)]$	_	_	3251s	1618m	1395m	1021w	618w	416w
$[Ni(HL_1)Cl_2(INH)]$	_	3404w	3219s	1606m	1413m	1060w	624w	406w
$[Cu(HL_1)Cl_2(INH)]$	_	_	3251s	1746m	1421m	1032w	623w	421w
$[Co(HL_2)Cl_2(INH)]$	3551b	3389s	_	1602m	1448m	1057w	640w	418w
$[Ni(HL_2)Cl_2(INH)]$	3408m	3275s	3076s	1608m	1437m	1062w	624w	410w
$[Cu(HL_2)Cl_2(INH)]$	3446s	3244s	3059s	1614m	1369m	1060w	574w	434w

Electronic Spectra and Magnetic Susceptibility Measurement

The electronic spectra measured in methanol and ethanol for the synthesized ligands and their metal complexes are presented in Table 3. In the UV spectra of HL_1 and HL_2 displayed one electronic transition each at 280 nm and 260 nm respectively and is assigned to the intraligand $\pi \rightarrow \pi$ * transition in the aromatic system (Arpi, 2003; Singh *et al.*, 2010). In the visible spectrum of the copper(II) complex of HL_2 one broad band around 620 nm due to d-d band and a slight shoulder at 800 nm as a result of distortion. This broadening may be due to Jahn-Teller effect arising from unequal occupation of the e_g pair of orbital (Singh *et al.*, 2010; Singh and Singh, 2012). The cobalt(II) complex of HL_2 showed a d-d transition band around 640 nm. In the ligand field spectrum of cobalt(II) complex of HL_2, two d-d bands (420 nm, 640 nm) are observed which are expected for the cobalt(II) ion in an octahedral environment corresponding to the transition $^3A_2g \rightarrow {}^3T_1g$ in the range 620-800 nm. Three d-d bands were also observed for Ni(II) complexes of HL_2 at 410, 548 and 930 nm as expected for nickel(II) ion in an octahedral environment (Cotton *et al.*, 2003; Akinyele *et al.*, 2019; Lee, 2005).

The visible spectrum of the Cu(II) complexes of the HL_1 showed two d-d bands observed at 630 nm and 820 nm respectively (Singh and Singh, 2012). The visible spectra of the cobalt (II) complex of HL_1 showed three d-d transitions around 585 nm, 620 nm and 980 nm respectively and the nickel(II) complexes of HL_1 showed two d-d transitions around 630 nm and 900nm respectively (Lee, 2005, Akinyele *et al.*, 2019).

The effective magnetic moments of the metal complexes are given in Table 3. The room temperature effective magnetic moment for the copper(II) complexes of HL_1 and HL_2 showed a magnetic moment of 2.08 BM and 1.84 BM respectively which are consistent for the d^9 electronic configuration with octahedral geometry (Akinyele *et al.*, 2020; Akinyele *et al.*, 2021). The observed values are due to ferromagnetism leading to high value of µeff. The magnetic susceptibilities of the synthesized cobalt(II) complexes are indicative of d^7

electronic configuration with three unpaired electrons. The values from 3.18-4.26 BM observed for the complexes are consistent with the d^7 electronic configuration with octahedral geometry (Woods and Odunola, 2011). The magnetic moment of 2.37–3.14 BM for nickel(II) complexes are consistent with those reported for octahedral geometry with d^8 electronic configuration having two unpaired electrons (Devi *et al.*, 2013). The proposed structure for the complexes is presented in Figure 1, with the metal centre coordinated to the nitrogen and oxygen of the ligands in the equatorial positions, and the chloride in the axial positions.

Table 3: The Electronic Spectra Magnetic Data of the Ligands and their Metal Complexes

Compounds	Intraligands Transition (nm)	Charge Transfer (nm)	Ligand Field Transition (nm)	Assignment	Proposed Structure	μ_{eff} (BM)
$C_6H_7N_3O$	257	_	_	_	_	-
HL_1	280	_	_		_	-
[Co(HL_1)Cl_2(INH)]	240, 300	405	585, 620, 980	${}^4T_{1g}\rightarrow{}^4T_{2g}$(P), ${}^4T_{1g}\rightarrow{}^4A_{2g}$ ${}^4T_{1g}\rightarrow{}^4T_{2g}$(F)	Octahedral	3.18
[Ni(HL_1)Cl_2(INH)]	380	405	630, 900	${}^3A_{2g}\rightarrow{}^3T_{1g}$(P), ${}^3A_{2g}\rightarrow{}^3T_{2g}$	Octahedral	2.41
[Cu(HL_1)Cl_2(INH)]	260	400	630, 890,	${}^2T_{2g}\leftarrow{}^2E_g$	Octahedral	1.78
HL_2	260	_	_			
[Co(HL_2)Cl_2(INH)]	350	420	640, 920	${}^4T_{1g}\rightarrow{}^4A_{2g}$ ${}^4T_{1g}\rightarrow{}^4T_{2g}$(F)	Octahedral	3.69
[Ni(HL_2)Cl_2(INH)]	305	410	548, 930	${}^3A_{2g}\rightarrow{}^3T_{1g}$(P), ${}^3A_{2g}\rightarrow{}^3T_{2g}$	Octahedral	3.05
[Cu(HL_2)Cl_2(INH)]	270, 298	405	620,800	${}^2T_{2g}\leftarrow{}^2E_g$	Octahedral	2.16

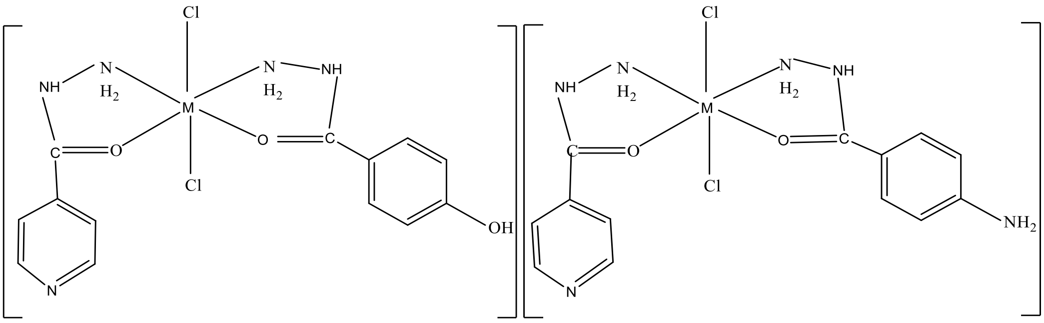


Figure 1: Proposed Structure for the Metal Complexes (M = Co, Ni or Cu)

Antimicrobial Activity

The antibacterial activities of the ligands and the metal complexes against different micro-organisms are presented in Table 4. The results showed that a reduced antibacterial activity was observed on coordination of the metal ions with the ligand. The ligands HL_1 and HL_2 exhibited better antibacterial activities compared with all their counterpart metal complexes. *S. aureus*, *Streptococcus spp* and *Salmonella spp* were susceptible to HL_1 and HL_2. All tested organisms were not susceptible to $[Cu(HL_1)Cl_2(INH)]$ and $[Co(HL_1)Cl_2(INH)]$ compared with the ligands HL_1 and HL_2. However, *Staphylococcus aureus* was susceptible to $[Ni(HL_1)Cl_2(INH)]$ with diameter of the zone of inhibition of 14mm. *E. coli* was susceptible to $[Co(HL_2)Cl_2(INH)]$ with diameter of zone of inhibition of 10mm. *Shigella spp* was also found to be susceptible to $[Cu(HL_2)Cl_2(INH)]$.

Table 4: Zone of Inhibition of Metal Complexes

Compounds	***S. aureus***[a]	***Strepto. spp***[a]	***B. cerus***[e]	***B.subtilis***[a]	***E. coli***[b]	***Salmo. spp***[b]	***Shigella spp***[b]
$[Cu(HL_1)Cl_2(INH)]$	_	_	_	_	_	_	_
$[Co(HL_1)Cl_2(INH)]$	_	_	_	_	_	_	_
$[Ni(HL_1)Cl_2(INH)]$	14	_	_	_	_	_	-
$[Cu(HL_2)Cl_2(INH)]$	-	_	_	_	_	_	8
$[Co(HL_2)Cl_2(INH)]$	_	_	_	_	10	_	_
$[Ni(HL_2)Cl_2(INH)]$	_	_	_	_	_	_	_
HL_1	14	20	_	-	8	16	14
HL_2	10	24	_	-	_	10	_
INH	_	_	_	_	_	-	_
10% DMSO	_	_	_	_	_	_	_
Streptomycin	20	_	22	14	22	24	20

“a” means gram-positive while “b” means gram-negative organisms

Diameter of Zone of Inhibition in mm of the Metal Complexes

Conclusions

Two hydrazides and their mixed ligand metal complexes with isoniazid have been prepared and characterized by physical and chemical methods. The infrared spectral showed that the coordination of the ligands to the metal ions is via the amino nitrogen ($-NH_2$) and carbonyl oxygen (C=O) suggesting that the ligands coordinate as bidentate ligands. The electronic spectra data supported octahedral geometry as proposed. This observation was supported by magnetic measurement. The synthesized compounds showed moderate activity against the tested microorganisms with the hydrazides displaying better antimicrobial activities.

References

Agwara, M. O., Ndifon, P. T., Ndosiri, N. B., Paboudam, A. G., Yufanyi, D. M., Mohamadou, A. (2010). Synthesis, Characterisation and Antimicrobial Activities of Cobalt(II), Copper(II) and Zinc(II) Mixed-Ligand Complexes Containing 1,10 Phenanthroline and 2,2-bipyridine. *Bulletin of the Chemical Society of Ethiopia, 24(3):383-389.*

Ainscough, E. W., Brodie, A. M., Dobbs, A. J. and Ranford, J. D. (1998). Antitumor copper(II) salicyaldehyde benzoyl hydrazone (H_2sb) complexes: Physicochemical Properties and the Single-crystal X-ray Structures of $[\{Cu(H_2sb)(CCl_3CO_2)_2\}_2]$ and $[\{Cu(Hsb)(ClO_4)(C_2H_5OH)\}_2]$ and the related salicylaldehyde acetylhydrazone (H_2sa) complex, $[Cu(Hsa)Cl(H_2O)]H_2O$. *Inorganic Chemistry. 267: 27-38.*

Akinyele, O. F., Akinnusi, T. O., Ajayeoba, T. A., Ayeni, A. O., Durosinmi, L. M. (2019) Synthesis, Characterization and Antimicrobial Activities of Cobalt(II), Nickel(II) and Copper(II) complexes of aroylhydrazone mixed with Aspirin, *Science Journal of Chemistry*, 7(3): 67-71.

Akinyele, O. F., Fakola, E. G., Durosinmi, L. M., Ajayeoba, T. A. and Ayeni, A. O. (2019). Synthesis and Characterization of heteroleptic Metal complexes of Isoniazid and Metformin. *Ife Journal of Science*, 21(3): 185-191.

Akinyele, O. F., Fakola, E. G., Durosinmi, L. M., Ajayeoba, T. A. and Ayeni, A. O. (2020) Synthesis, Characterization and Antimicrobial Activities of heteroleptic Metal Chelates of Isoniazid and 2,2'-bipyridine. *Bull. Chem. Soc. Ethiop*. 34(3), 471-478.

Akinyele, O. F., Fakola, E. G., George, R. C., Durosinmi, L. M. (2021). Synthesis, Characterization and Antibacterial Activity of mixed ligand complexes of nicotinamide and 2,2'-bipyridine. *Ife Journal of Science,* 23(1): 213 - 222.

Al-Shaalan, N.H., (2007). Antimicrobial Activity and Spectral, Magnetic and Thermal studies of some Transition Metal Complexes of a Schiff base hydrazone containing a quinoline moiety. *Molecules*, 12(5), pp. 1080-1091.

Al-Shaalan, N. H. (2011). Synthesis, characterization and biological activities of Cu(II), Co(II), Mn(II), Fe(II) and UO_2(VI) complexes with a new Schiff base hydrazone: O-hydroxyaceto- phenone-7-chloro-4-quinoline hydrazone. *Molecules*, 16: 8629-8645.

Anwar, M. U., Elliott, A.S., Thompson, L.K. and Dawe, L.N., (2011). Complexes of functionally modified hydrazone ligands-square, rectangular (M4), trigonal bipyramidal (M 5) and pin-wheel (M6) motifs. *Dalton Transactions*, 40(17), pp. 4623-4635.

Arpi, M., (2003). Six cases of *lactobacillus bacteraemia*: Identification of Organisms and Antibiotic Susceptibility and Therapy. *Scandinavian Journal of Infectious Diseases*. 35:404Y8.

Chohan, Z. H. and Kauser, S. (2000), Biological Activities of Hydrazone Derivatives," *Molecules,* Vol. 12, No. 8, (2007), pp. 1910-1939.

Chohan, Z. H., Arif M., Shafiq Z., Yaqup M., Supuran C.T., (2006). Complexes of Mn(II), Fe(II), Co(II), Ni(II), Cu(II), Zn(II) with a bishydrazone *synthesis and reactivity in Inorganic, Metal-Organic and Nano-Metal Chemistry* 35: 837-844.

Cotton, F. A.; Wilkinson, G., Murillo, C.A., Bochmann, M., (2003). Advanced Inorganic Chemistry, 6th Ed. John Wiley and Sons Inc., New York.

Deepa, K. P. and Aravindakshan, K. K. (2005). Synthesis, Characterization and Antifungal Studies of Metal Complexes of Benzoyl and Salicylhydrazones of Bromoacetanilide. *Synthesis and Reactivity in Inorganic, Metal-Orgain and Nano-Metal Chemistry,* 35: 409-416.

Devi, G. S., Muthu, A. K., Kumar, D. S., Rekha, S. Indhumathi, R. and Nandhini, R. (2013) "Studies on the antibacterial and antifungal activities of the ethanolic extracts of *Luffa cylindrica* (Linn) fruit," *International Journal of Drug Development and Research*, 1, 105–109.

Geary, W. J., (1971). The Use of Conductivity Measurements in Organic Solvents for Characterization of Coordination Compounds. *Journal of Coordination Chemistry Reviews*.7, 81-83.

Howlader, M. B. H., Tarafder, M.T.H and Islam, M. A. (2009). Palladium(II) Complexes of Hydrazones derived from 4-dodecyloxybenzoylhydrazine with some Aliphatic and Aromatic aldehydes and their mesogenic behavior; *Indian Journal of Chemistry,* 48A; 1078-1084.

Lee J. D. *Concise Inorganic Chemistry*.5th Edition. India: Blackwell Science Limited. 2005, 205 -324.

Lashanizadegan, M and Jamshidbeigi, M: (2012) Synthesis of n, n'-bis[4-(benzeneazo) salicylaldehyde] 4- methyl-1-2-phenylenediamine and its transition metal complexes. *Synthesis and Reactivity in Inorganic, Metal-Organic, and Nano-Metal Chemistry*. 42: 507-512.

Mitu, L., Ilis, M., Raman, N., Imran, M and Ravichandran, S. (2012) Transition metal complexes of isonicotinoylhydrazone-4-diphenylaminobenzaldehyde: Synthesis, Characterization and Antimicrobial Studies. *E-Journal of Chemistry,* 9: 365-372.

Mostafa, M., (2011) Metal Chelates of Hydrazone Ligand Chelating Tendencies of 2-Carboxy Phenylhydrazoacetoacetanilide (2-Cphaa) Ligand, *Research Journal of Chemical Science*.,1(7), 1-4.

Nakamoto K., (2009). Infrared and Raman Spectra of Inorganic and Coordination Compounds: Applications in Coordination, Organometallic, and Bioinorganic Chemistry, sixth ed., Wiley-Blackwell, 79:7860-7867.

Singh, D. P., Malik, V., Kumar, K., Sharma, C., Aneja, K.R. (2010): Macrocyclic Metal Complexes derived from 2,6-diaminopyridine and Isatin with their Antibacterial and Spectroscopic Studies. *Spectrochimical Acta* (A), 76:45-49.

Singh, J. and Singh, P. (2012). Synthesis, Spectroscopic, Characterization, and in-vitro Antimicrobial Studies of pyridine-2-carboxylic acid N-(4-Chloro-Benzoyl)-hydrazide and its Co(II), Ni(II), and Cu(II) complexes. *Bioinorganic Chemistry and Applications*, Article ID 729708.

Singh, H. L and Varshney, A. K. (2006) Synthetic, Structural, and Biochemical Studies of Organotin(IV) With Schiff Bases Having Nitrogen and Sulphur Donor Ligands, *Bioinorganic Chemistry and Applications*, Article ID 23245, Pages 1–7.

Thaker, B. T., Modi C. K., Ashwin S. and Patel, A. S. (2005). Synthesis, Spectra, Magnetic and Thermal Studies of the Complexes of Co(II) and Ni(II) with some Bidentate and Tridentate hydrazone Ligands. *E-journal of Chemistry* 2.6: PP; 21-29.

Woods, J. A. O. and Odunola, O. A. (2011). New Ni(II) Mixed Ligand Complexes of Dithiocarbamates with Schiff base. *Journal of Chemical and Pharmaceutical Research 3.6:865-871.*