ISSN: 0975 -8542

Journal of Global Pharma Technology

Available Online at: www.jgpt.co.in

RESEARCH ARTICLE

Synthesis and Characterization of Azo Dyes Ligands Complexes with Rh (III) and La (III) and Studies Their Industrial and Bacterial Application

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Abstract

1-[4-(2-Hydroxy-4, 6-dimethyl-phenylazo)-phenol]-ethanone (HL_1) and 2-(4-methoxy-phenylazo)-3, 5-dimethyl-phenol (HL_2) were produced by combination the diazonium salts of amines with 3, 5-dimethylphenol. The geometry of azo compounds was resolved on the basis of (C.H.N) analyses, ^1H and $^{13}CNMR$, FT-IR and UV-Vis spectroscopic mechanisms. Complexes of La (III) and Rh (III) have been performed and depicted. The formation of complexes has been identified by using elemental analysis, FT-IR and UV-Vis spectroscopic process as well, conductivity molar quantifications. Nature of complexes produced have been studied obeyed mole ratio and continuous alteration ways, Beer's law followed through a concentration scope (1×10^{-4} - 3×10^{-4} M). High molar absorbtivity from the compound solutions were noticed. Analytical data showed that all the complexes offered 1:3 metal-ligand ratios. In the radix from physicochemical datum an octahedral structure was described of the complexes. Biological activity of these compounds was assayed. Other than, the dyeing carried out of the produced compounds was practical in cotton fabric. The dyes were checked into light and detergent firmness.

Keywords: *Complexes, Azo dyes, Biological activity, Acetophenone, Aniline.*

Introduction

Azo dyes are very significant molecules and have many applying in cases of industrial and coordination chemistry [1, 2]. Azo dyes are identified by the attendance of the azo half (-N=N-) in their edifice connected with two aromatic or aliphatic groups. Because of their fixed physicochemical estates and biological activity, they have found a wide applying in pharmaceutical, cosmetic, food, dyeing textile industry and analytical chemistry [3].

Azo molecules are the oldest and largest species of industrial synthesized organic dyes lead to their more applying in varied field [4, 6]. The complexes of azo dyes ligands with transition metals are of present attraction describe to the used physical, chemical, photo physical and photochemical, catalytic and differing material estates [7, 9]. On that work, ligands of azo functional group obtained of 4-methoxyaniline and 4-aminoacetophenone like diazo component as well 3, 5-dimethylphenol like coupling agent, were produced. The complexes from Rh (III) and La (III) with these ligands have been produced and identified physcio-chemically.

Experimental

Instrumentation

Atomic absorption was registered through employing Shimadzu A.A-160A Atomic Absorption/Flame Emission Spectrophotometer. The ^{13}C and ^1H-NMR spectrum were noted at Brucker-300 MHz Ultra Shield spectrometer in Al- al- Bayt University utilizing DMSO as solvent and TMS as reference. (C, H, N) analysis were well-done on Al- al- Bayt University, Jordan, employing Euro vector EA 3000A Elemental Analyzer.

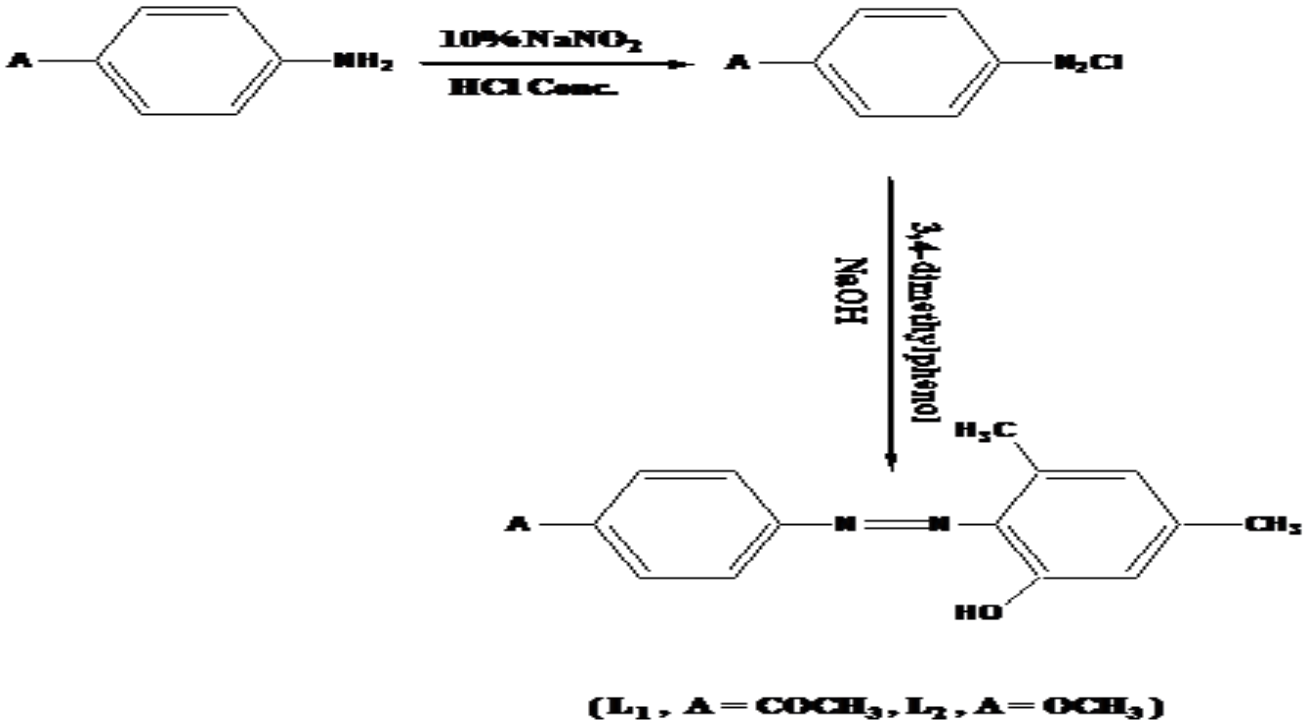
Conductivity of the complexes dissolved in ethanol (10^{-3} M) was recorded at 25°C using Philips PW- Digital Conductometer. UV- Vis spectrum was recorded by a Shimadzu UV-160A Ultra Violet-Visible Spectrophotometer.IR- spectra were possessed at Shimadzu, FT-IR- 8400S Fourier Transform Infrared Spectrophotometer within 4000- 400 cm^{-1} spectral areas for samples produced like KBr discs. Another that than, melting points were complete utilizing Stuart Melting Point Apparatus.

Materials and Reagents

Obedience chemicals were utilized as collected of provider: Lanthanum chloride nonahydrate (98.8%) and Rhodium chloride monohydrate (98.8%) (Merck) , 4-methoxyaniline, 3, 5-dimethylphenol and 4-aminoacetophenone (B.D.H).

Preparation to the ligands

A solution was produced [10] of amines (0.342 gm and 0.307 gm, 1mmole) in (10ml) of EtOH obtaining (2ml) conc. HCl which was diluted for 10 ml H_2O, as well diazotized in 5°C for 10% solution from $NaNO_2$. The diazotized solution was added drop of wisdom for stirring into cooled ethanolic solution for (0.305 gm, 1mmole) to the 3, 5-dimethylphenol. Then 25 ml for1M NaOH solution was followed into dark- colored mixture and precipitation for azo ligand was noticed. This precipitate was filtrated, washed numbering ounces for (1:1) ethanol: water, then leave the mixture to dry. The interaction appears in Scheme 1, while Table 1 characterizes the physical estates and elemental analysis.



Scheme-1: Synthesis of the azo ligands

Table-1: Physical properties for the ligands and complexes

Compounds	Color	m.p°C	Yield%	Analysis Calc (Found)			
				M%	C%	H%	N%
Ligand(HL_1)	Orange	186	81	-	71.64 (70.69)	5.97 (4.97)	10.44 (9.87)
[Rh(L_1)$_3$]	Brown	271	75	11.39 (10.68)	63.71 (62.99)	4.97 (4.36)	9.29 (8.89)
[La(L_1)$_3$]	Reddish orange	223	77	14.78 (13.88)	61.27 (60.83)	4.78 (3.97)	8.93 (7.75)
Ligand(HL_2)	Yellow	148	74	-	70.31 (69.84)	6.25 (5.93)	10.93 (9.93)
[RhL_2)$_3$]	Brown	185	70	11.86 (10.68)	62.21 (61.82)	5.18 (4.76)	9.67 (8.75)
[La(L_2)$_3$]	Reddish brown	210	73	15.37 (14.64)	59.73 (58.77)	4.97 (4.33)	9.29 (8.85)

Buffer Solution

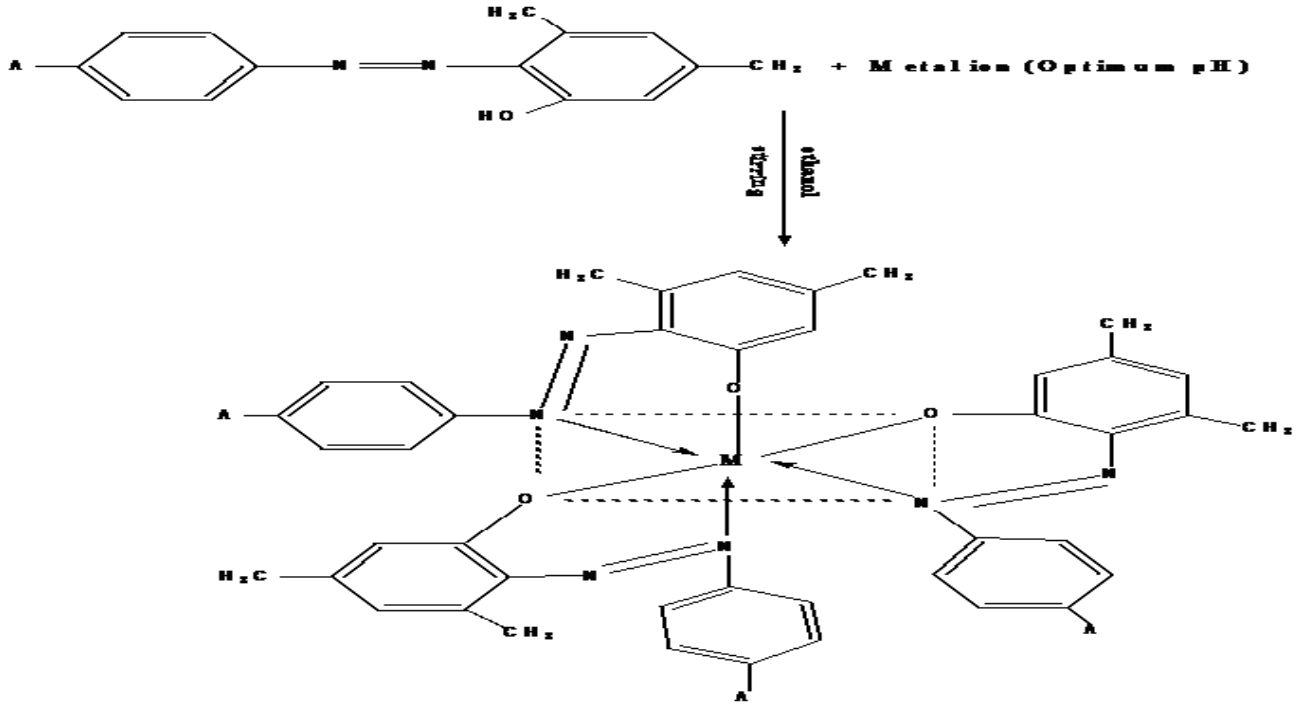
(0.01M, 0.771 gm) from ammonium acetate was resolved at one liter from doubly deionized water. At only pH medium (3-9) was employment acetic acid or ammonia solution.

The Standard Solution

A block of standard solutions to the $LaCl_3.9H_2O$ and $RhCl_3.H_2O$ were made in varying concentration (10^{-5}-10^{-3} M) on pH range (3-9). On the same time a block from ethanolic solutions for ligands during the range of concentrations (10^{-5}-10^{-3} M) was likewise prepared.

Preparation for Metal Complexes (Public Method)

Ethanolic solution for the ligands (0.268gm and 0.256gm, 3mmole) was added drop wise for stirring into the 0.117gm and 0.075gm for $LaCl_3.9H_2O$ and $RhCl_3.H_2O$ resolved at pH solution for the required pH. The mix was cooled to dark color precipitate was gained, filtrated, and washed number ounces for 1:1 water: ethanol mix, thereafter for acetone. The preparation method is shown in Scheme-2.



L_1, A = $COCH_3$, L_2, A = OCH_3, M = La(III) and Rh(III)

Scheme- 2: The expected structure of the metal (III) complexes of (L_1 and L_2)

Study of Biological Activity

The antibacterial activity for these compounds was determined by using appreciation similar with the traditional disc diffusion method [11]; sterile 5 mm filter paper discs (Whatman, No.1) were soaked in this compound (Disc loaded with the DMSO as solvent and like control) and permitted complete evaporation to be used. Then discs were placed onto the surface of the Muller Hinton agar plates at different areas on the surface of each plate, after a 24 hr culture of the pathogenic strains (*E. coli, S. aureus and B. cereus*) were spreading over the surface of Muller Hinton agar plates with a sterile cotton swab. The plates were incubated at 37C° for 24 hr. The Results were specified through registration the diameter (mm) to a zone from inhibition around every disc at the plate.

Dyeing Method

The dyeing estates from the generated compounds were tasted and used during cotton fabric like (1% shade). Fabric dye was formative in (15- 20C°) at (1 hr), and in pH (10).

Results and Discussion

For the preparing of the ligands (HL_1 and HL_2) a linking from 3, 5-dimethylphenol for the appropriate diazotized at alkaline solution was performance. Synthesized ligands were identified by (C, H, N) analysis,

^{1}H and ^{13}CNMR, FT-IR, and UV-Vis spectral. Aqueous-ethanolic solutions were constantly acquired into survey the interaction from the metal ions La (III) and Rh (III) for the produced ligands. The Colors from these mixed solutions through the molar condensation and acidity area perfect were various of brown into orange.

NMR Spectrum

The ^{1}HNMR spectrum of the ligand (HL_1) at DMSO (Figure-1) displays various gestures in δ=6.623-8.135 ppm attributed into aromatic protons [12], the gesture at δ=10.130 ppm lead to proton of phenol [13]. The resonance at δ=2.248-2.345 ppm is assigned to δ (CH_3) groups and the gesture at δ=2.50 ppm pointed into DMSO-d_6 [14]. The ^{13}CNMR spectrum of the (HL_1) (Figure-2) display gestures at δ=26.889 ppm and δ=20.522 ppm due to carbon of (CH_3) in acetyl and phenol groups. The resonance at δ=197.359 ppm assigned to carbonyl band. The various gestures at (δ=159.395, 137.241, 136.302, 129.500, 128.396, 121.799 and 116.227 ppm attributed to carbon atoms of aromatic rings. The symbol at δ= 142.208 ppm due to carbon of (-OH) group and the indicative in δ=39.471 ppm due into DMSO-d_6 [15].

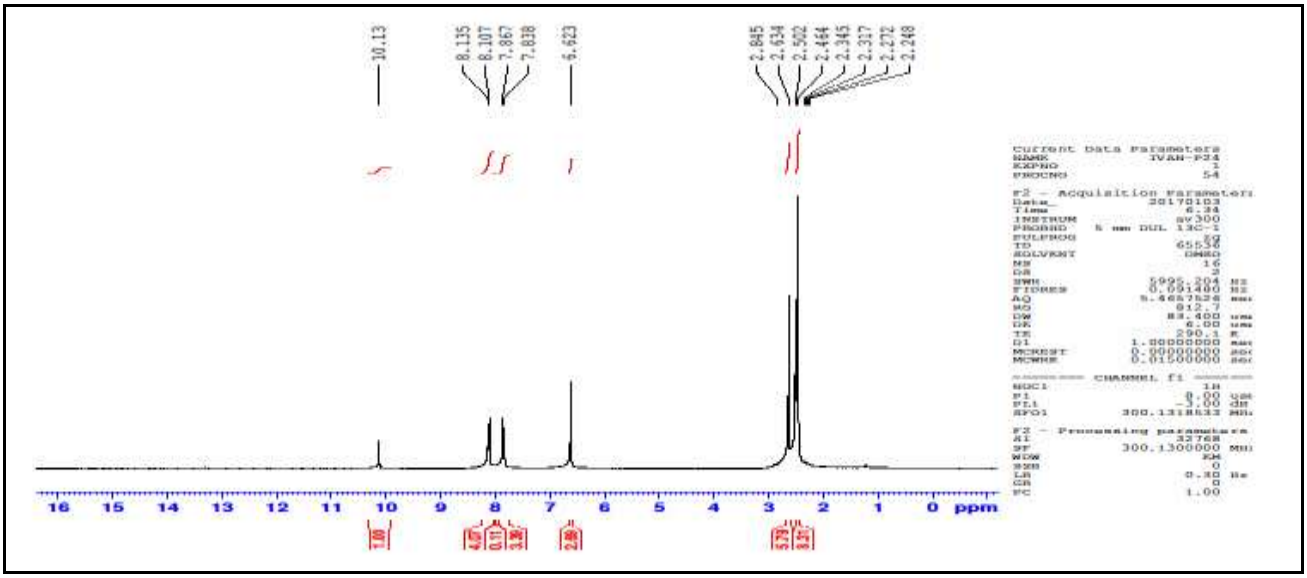


Fig.1: ^{1}HNMR spectrum of the ligand (HL_1)

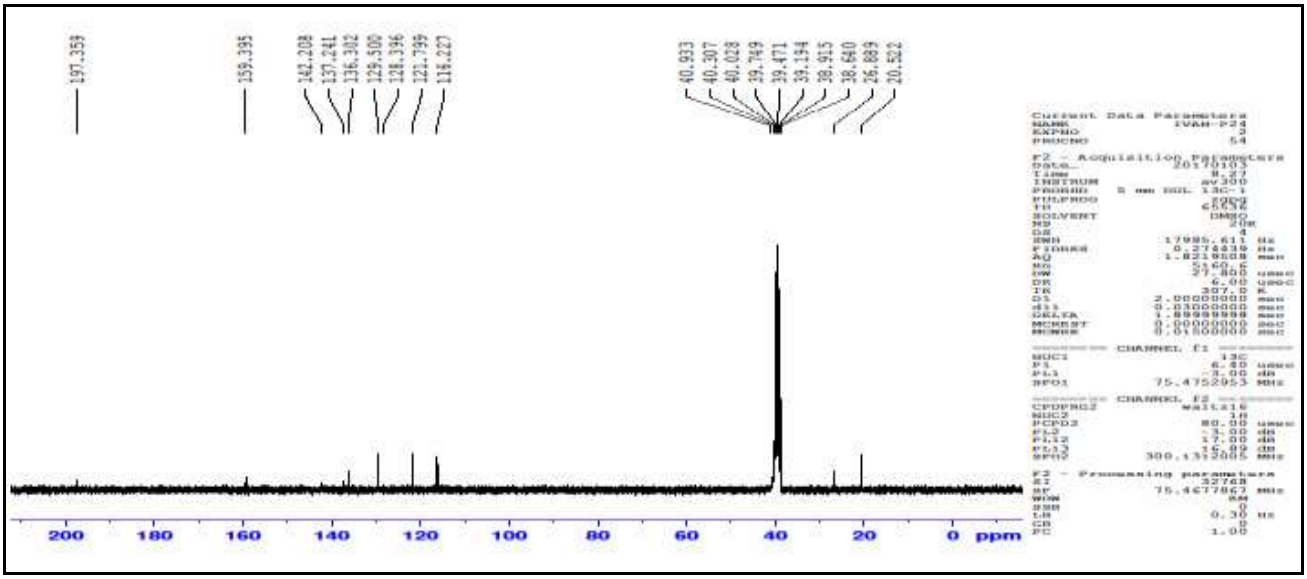


Fig.2: ^{13}CNMR spectrum of the ligand (HL_1)

The ^{1}HNMR spectrum from the ligand (HL_2) (Figure-3) shows various gestures in δ=6.299-8.135 ppm described into aromatic protons [16]. The symbol at δ=9.782 ppm lead into proton for phenol [17]. The symbol in δ=3.845 ppm due into δ (CH_3) of methoxy group, and the signals in δ=2.184 and δ=2.368 ppm attributed into δ (CH_3) of phenol group. On the other hand, the symbol in δ=2.5 ppm due into DMSO-d_6 [18]. The ^{13}CNMR spectrum for the (HL_2) (Figure-4) display various gestures in (δ=161.050, 146.952, 142.398, 134.280,

123.465, 115.864 and 114.384) ppm described into carbon atoms from aromatic rings. The signal in δ=157.729 ppm lead into carbon for (-OH) group. The symbol at δ= 55.515 ppm due into carbon for (CH_3-O-). The signals at δ=19.940 and 17.529 ppm attributed to δ (CH_3) groups of phenol. The gesture at δ=39.476 ppm due to DMSO-d6 [15].

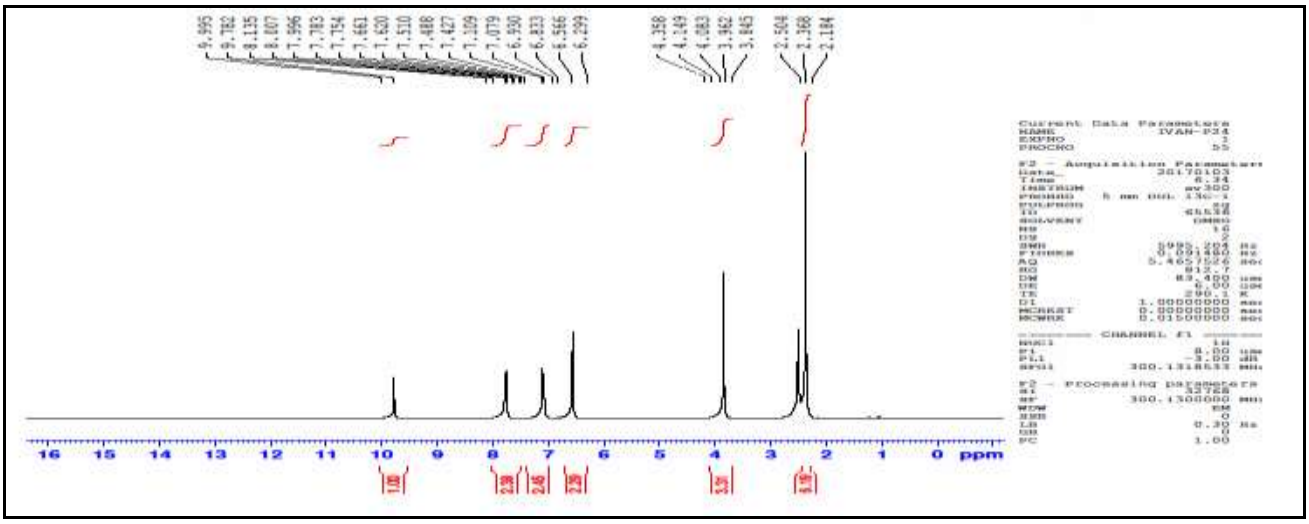


Fig.3: ^{1}HNMR spectrum to the ligand (HL_2)

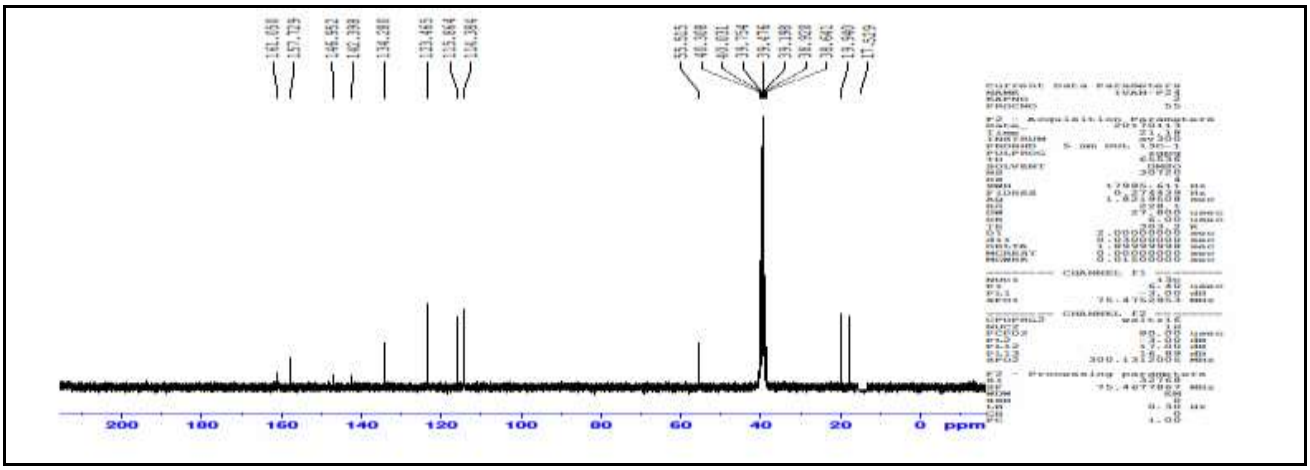


Fig.4: ^{13}CNMR spectrum of the ligand (HL_1)

Calibration Curve

Various molar focus (10^{-5}–10^{-3} M) from mixed aqueous-ethanolic for ligand and metal ions, only the concentration at the extent (1-3×10^{-4}M) pursued Beer's law and showed evident intensive color. Better fit right lines were occurred for correlation factor R>0.9980 like shown at Figure-5.

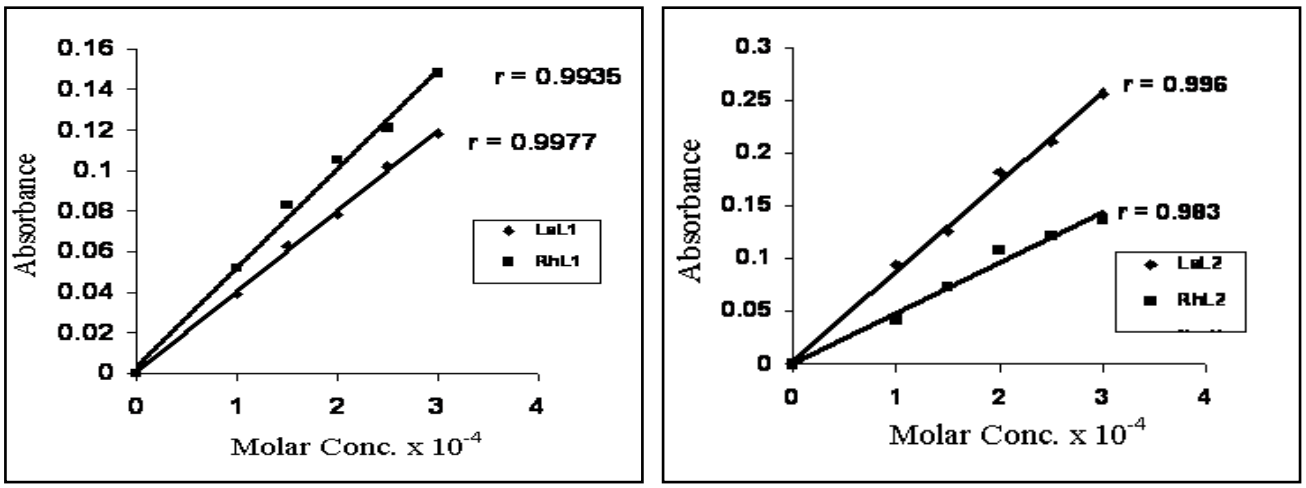


Fig.5: A linear relationship between molar concentration and absorption

Optimum Conditions

At search out the interaction amidst the generated ligand and metal ions under leaching to the production from the compounds, the spectrum to merging solutions to the ligand and metal ions into arrive for optimum pH and focus, as well firm wave length (λ_{max}) were studies firstly. After that mole ratio metal to ligand (M: L) was knew into equip the compounds. Perfect focus was selection to compound solution based on whom solution affords the highest absorbance in constant (λ_{max}) in various pH, and consequence are labeled in Table-2. The exam consequence proofs in order to the absorbance from whole intended compounds are maximum and constant at a buffer solution for ammonium acetate at the pH scope (3-9). It was found that whole prepared compounds had exemplary pH like is shown at Figure-6.

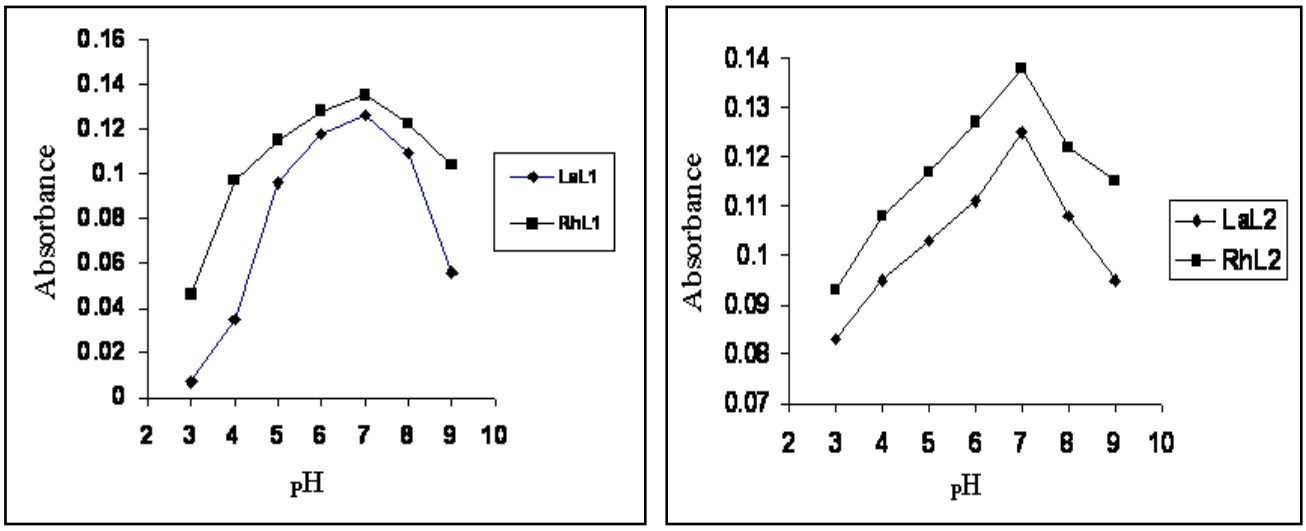


Fig.6: Influence for pH at absorbance (λ_{max}) of compounds

Stoichiometry of Compounds

The typesetting from compounds shaped at solutions has been appointed through mole ratio and job manners. At both situations the consequences spread a 1:3 (metal: ligand) ratio. The selected plot is shown at Figure-7. Table-2 synopsizes the outcomes gated, as well specification to the making compounds

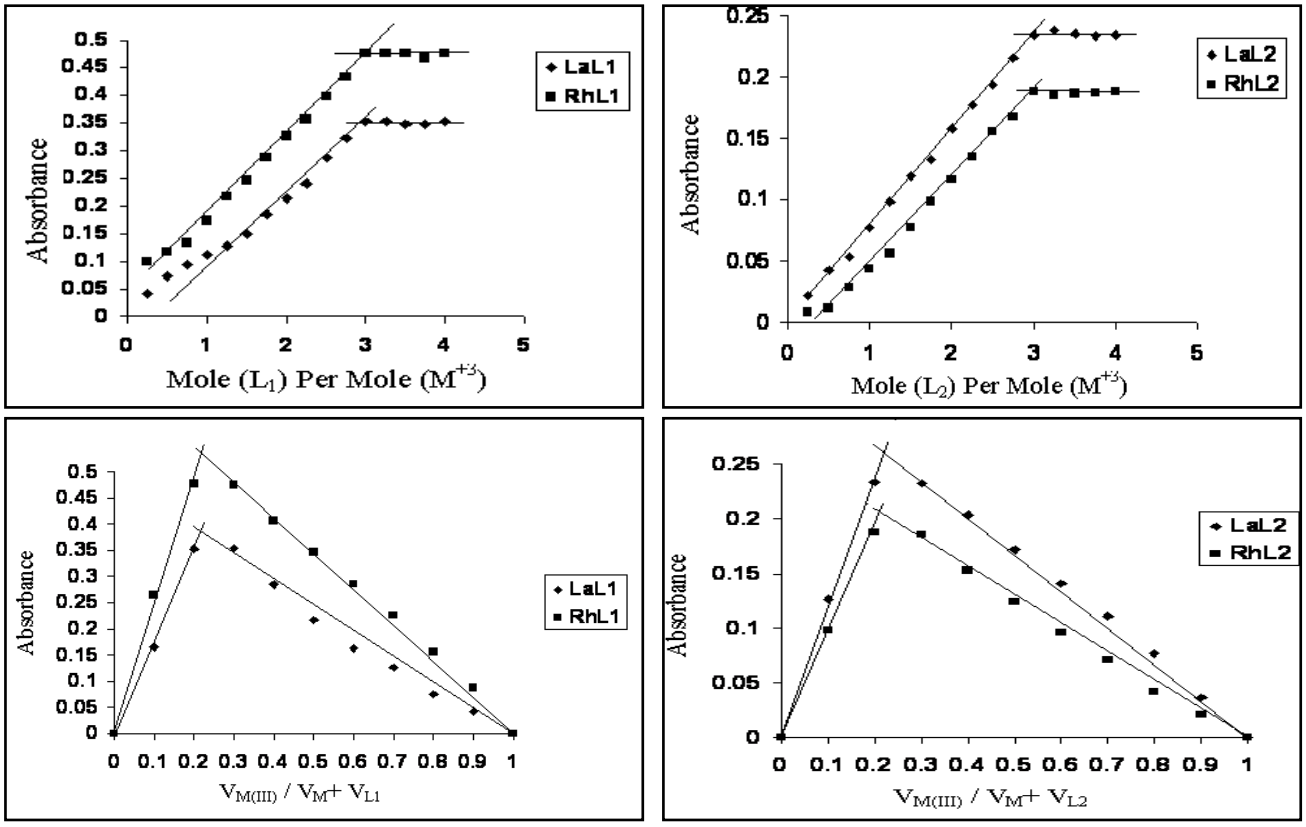


Fig.7: Mole ratio and Job manners into compounds· solutions

Physical Properties

The solid compounds have been produced through immediate interaction to the ligand melted in ethanol for the metal ions melted within perfect pH and at a (Metal: Ligand) ratio of 1:3. The outcome of elemental analyses and the metal import from these compounds were too real identical for the studied values. The molar conductance from the complexes (10^{-3} M) melted in ethanol display those non- electrolytic nature [19] datum are included at Table-2.

Table-2: Conditions to the preparation from the compounds, UV-Vis and conductance mensuration datum.

Compounds	Optimum pH	Optimum Molar Conc. x 10^{-4}	M:L Ratio	(λ_{max}) nm	ABS	ϵ_{max} (L.mol^{-1}.cm^{-1})	Λ_m(S.cm^2.mol^{-1}) In Absolute ethanol
Ligand(HL_1)	-	-	-	264 364 464	0.508 1.238 0.296	508 1238 296	-
[Rh(L_1)$_3$]	7	2.5	1:3	262 372 490 984	0.235 0.454 0.082 0.006	235 454 82 6	7.35
[La(L_1)$_3$]	7	2.5	1:3	264 372 493 890 984	0.357 0.754 0.077 0.009 0.019	357 754 77 9 19	11.71
Ligand(HL_2)	-	-	-	238 356 444	0.651 1.436 0.137	651 1436 137	-
[Rh(L_2)$_3$]	7	2	1:3	240 356 477 890 984	0.309 0.485 0.062 0.002 0.008	309 485 62 2 8	10.57
[La(L_2)$_3$]	7	2	1:3	244 356 498 892 984	0.212 0.432 0.059 0.006 0.013	212 432 59 6 13	11.76

Electronic Spectra

The UV-Vis spectrum from the produced compounds melted at ethanol (10^{-3} M) have been gauging and the datum received are recorded at Table 2. The UV- Vis spectra from ethanolic solution for the ligands 10^{-3} M (Figure-8 and 9) displayed generally three peaks, the first and second peaks in the average (238-364 nm) were attributed into the mild energy π- π* transition. The third peak in the average (444-464 nm) was gestured to the n- π* transition [20, 21].

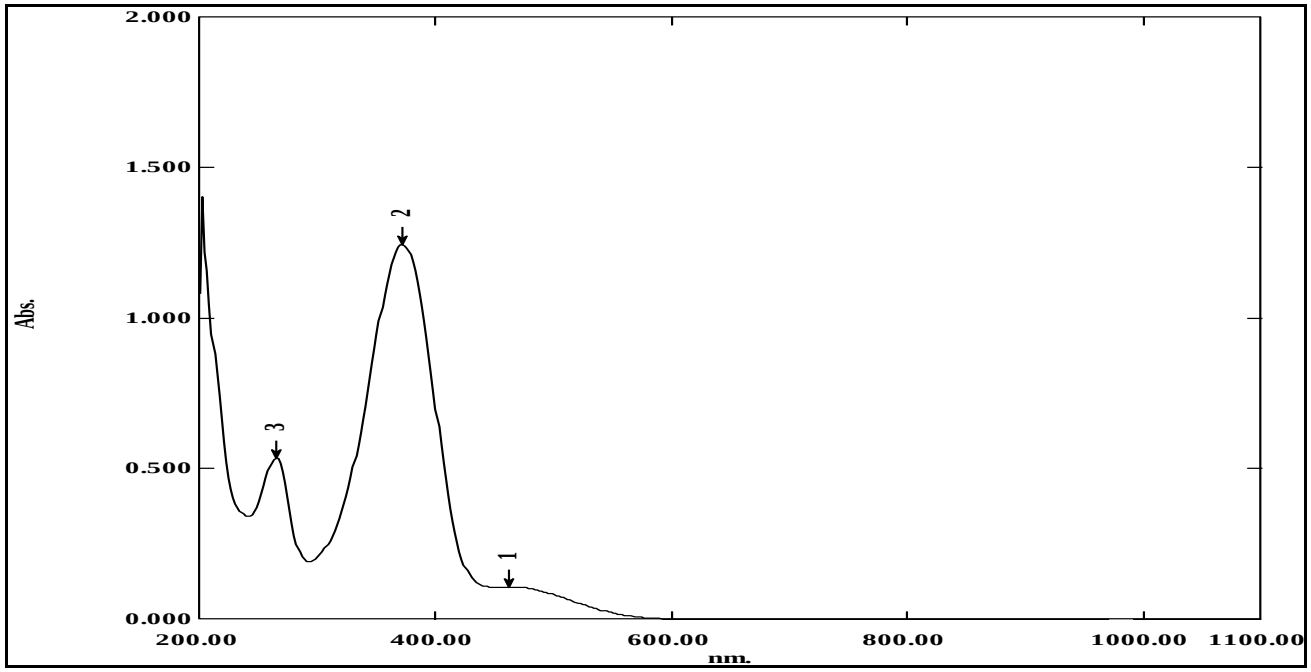


Fig.-8: UV-Vis spectroscopy to the ligand (HL_1)

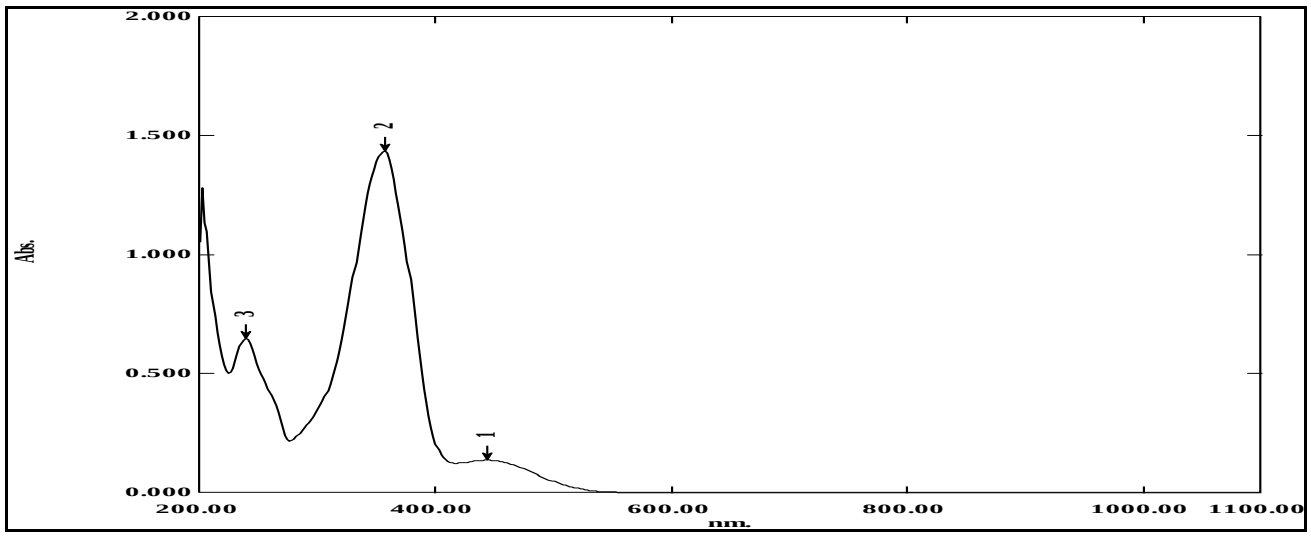


Fig.-9: UV-Vis spectroscopy to the ligand (HL_2)

The spectra of Rh(III) complexes (Figure-10 and 11) appeared absorption peaks in the average (240-372 nm) were concerning to ligand felid, after that another peaks in 477 and 490 nm were assigned to charge transfer. The spectra of these complexes show absorbance peaks at the average (890-984 nm) due into (d-d) electronic transition.

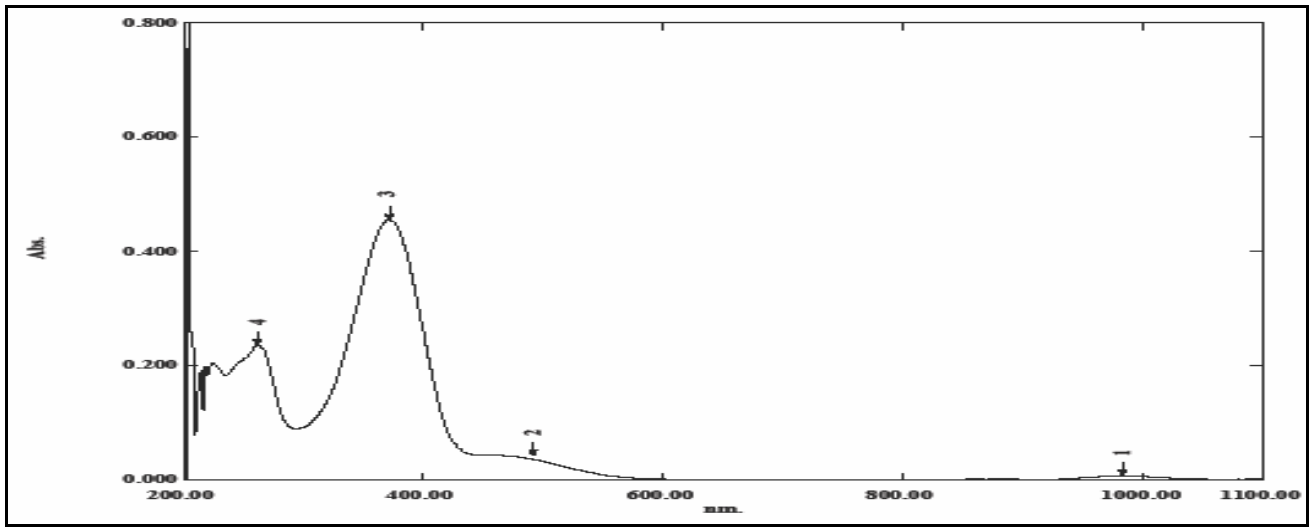


Fig.10: UV-Vis spectroscopy to the $[Rh(L_1)_3]$ complex

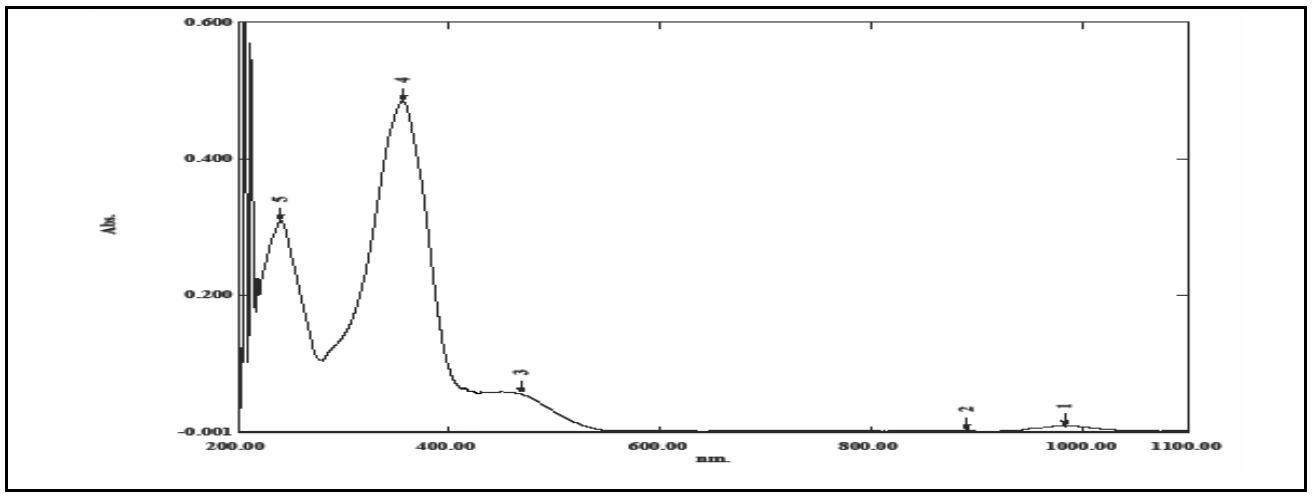


Fig.-11: UV-Vis spectroscopy to the $[Rh\ (L_2)_3]$ complex

The spectra of La (III) complexes (Figure-12 and 13) showed peaks at the average (244-372 nm) due to ligand felid. Other two peaks in 493 and 498 nm found to be caused by charge transfer. The peaks at the average (890-984 nm) due to (f-f) electronic transition [22].

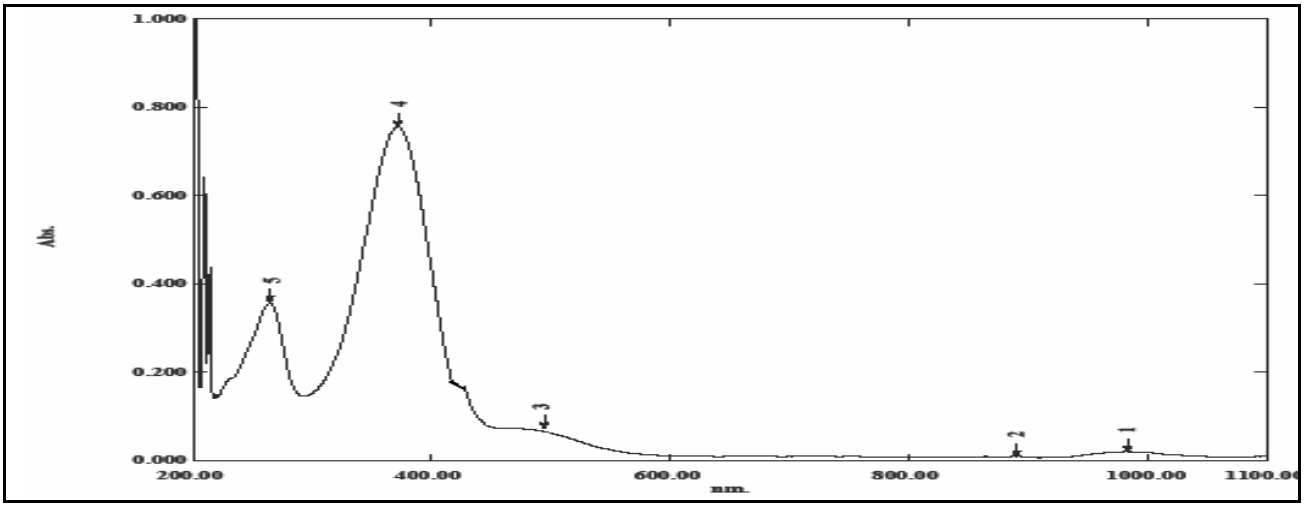


Fig.-12: UV-Vis spectroscopy to the [La $(L_1)_3$] complex

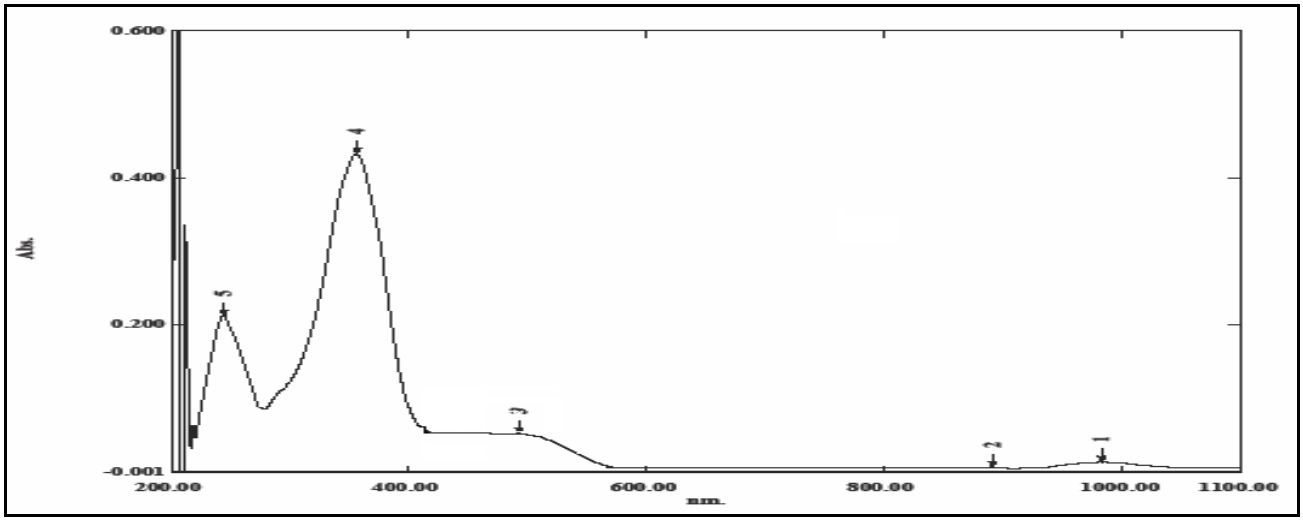


Fig.-13: UV-Vis spectroscopy to the [La $(L_2)_3$] complex

Infrared Spectra

The FT-IR spectrums from the produced compounds have been collated, and the data was scheduled in Table 3. The broad band in the FT-IR spectra of the ligands at 3360 and 3402 cm^{-1}, which was related into the stretching vibration for υ (OH) phenol, the disappearance of this band at the spectrum from whole produced compounds pointed out the deprotonation from phenol group to coordination with metal ion [23, 24]. The (HL_1) spectrum presented band at 1666 cm^{-1} which was attributed to the stretching of υ(C=O), ever after no significant variation at this band was observed, the possibility that coordination occur by the donating atom in this group was ruled out [25]. Bands differentiating of the azo groups at 1473 and 1504 cm^{-1} displaced to higher and lower wave number with alter in shape in spectra of all produced complexes [26, 27]. The bands in IR spectra of the ligands at the range (1350-1631 cm^{-1}) due to bending frequency of (δCH_3) and stretching vibration of υ(C=C) [28]. Stretching frequency bands for metal-nitrogen and metal-oxygen further [29, 30] proven through the presence from the bands about 447-509 cm^{-1}. Pursuant to the results protected, an octahedral geometry has been offered for the produced complexes.

Table-3: The major hesitancy to the ligands and compounds (cm^{-1})

Compounds	υ(OH)	υ(C=O) + υ(C=C)	υ (N=N)	$\delta CH_{3\ as,s}$	υ(M-N) + υ(M-O)
Ligand(HL_1)	3360 br.	1666 sh. 1600 s. 1577 sho.	1473 sh.	1435 s. 1400 sh 1354 sh. 1315 sh..	-
[Rh$(L_1)_3$]	-	1666 sh. 1600 s. 1577 sho.	1492 sh.	1435 sho. 1400 sh. 1354 sh. 1315 sh.	474 w. 447 w.
[La$(L_1)_3$]	-	1666 sh.	1492 s.	1435 sho.	509 w.

		1600 s. 1577 sho.		1400 sh. 1354 sh. 1315 sh.	466 w.
Ligand(HL_2)	3402 br.	- 1666 sh. 1597 sh.	1502 sh.	1438 sho. 1381 sho. 1346 sho. 1315 sh.	-
$[RhL_2)_3]$	-	- 1662 sh. 1597 sh.	1465 s.	1438 sho. 1381 sho. 1346 sho. 1315 sh.	528 w. 501 w.
$[La(L_2)_3]$	-	- 1597s.	1465 s.	1438 sho. 1381 sho. 1342 sho. 1315 sh.	528 w. 509 w.

Br=broad, sh=sharp, s=strong, sho=shoulder, w=weak

Biological Efficiency and Dyeing Properties

All the ready ligands and its complexes have been examined with Gram-negative and Gram-positive bacteria. Table-4 suggests the suppression spread converse the bacteria paradigm. The dyeing performance of the prepared compounds was defined at cotton fabric. The dyes were essay to the light and detergent constancy. So whole dyes appeared the dyeing is highly excellent holding and depth on the fabric. The dyeing was referred on Figure-14 and 15.

Table-4: Diameters (mm) fron suppression for bacteria to the ligands and complexes.

Compounds	Bacillus cereus	Staphylococcus aureus	Esherichia coli
Ligand(HL_1)	14	19	21
$[Rh(L_1)_3]$	16	20	25
$[La(L_1)_3]$	17	15	24
Ligand(HL_2)	10	18	25
$[RhL_2)_3]$	12	20	20
$[La(L_2)_3]$	17	15	23

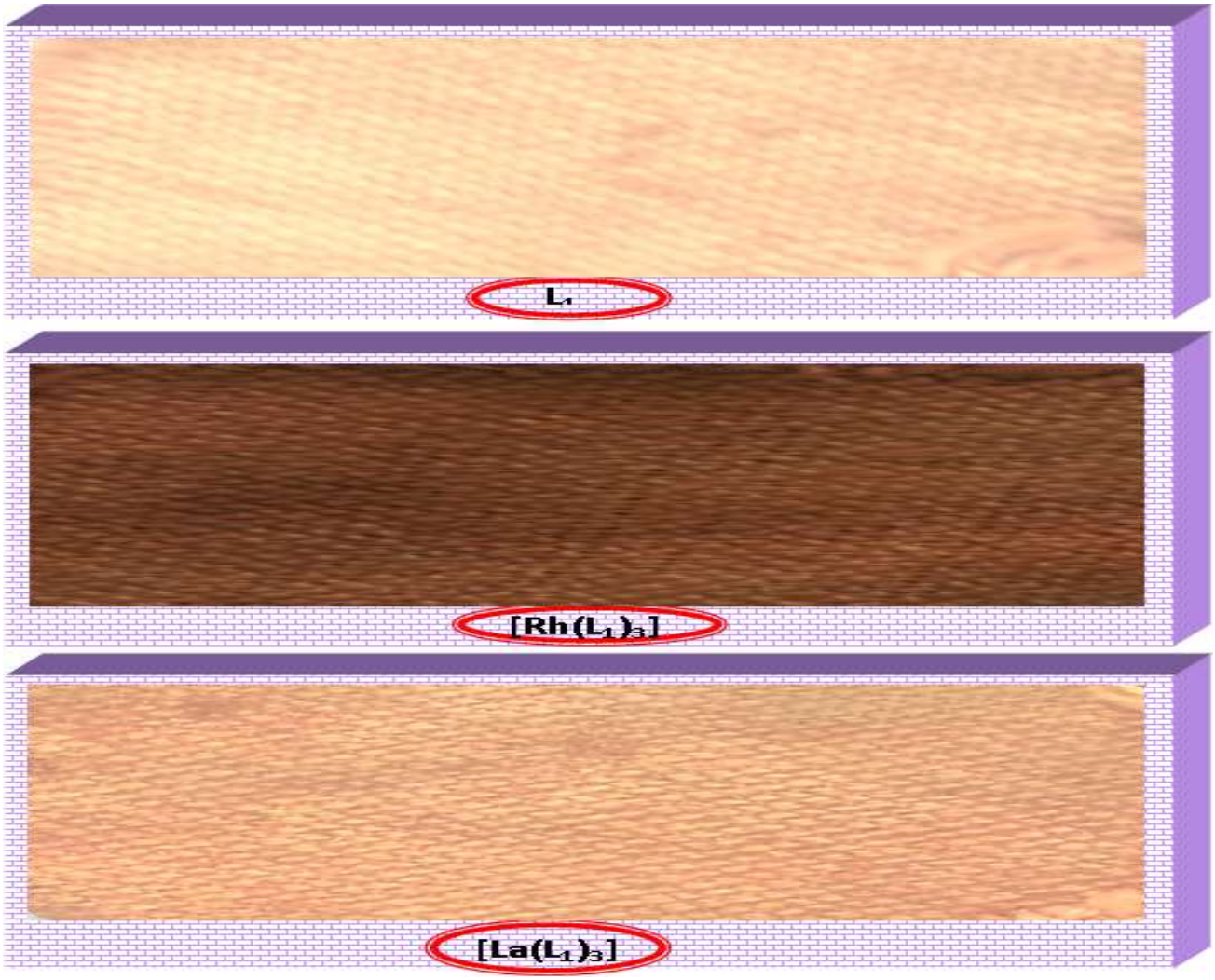


Fig.14: Samples the textiles dyeing of the ligand (HL_1) and their complexes

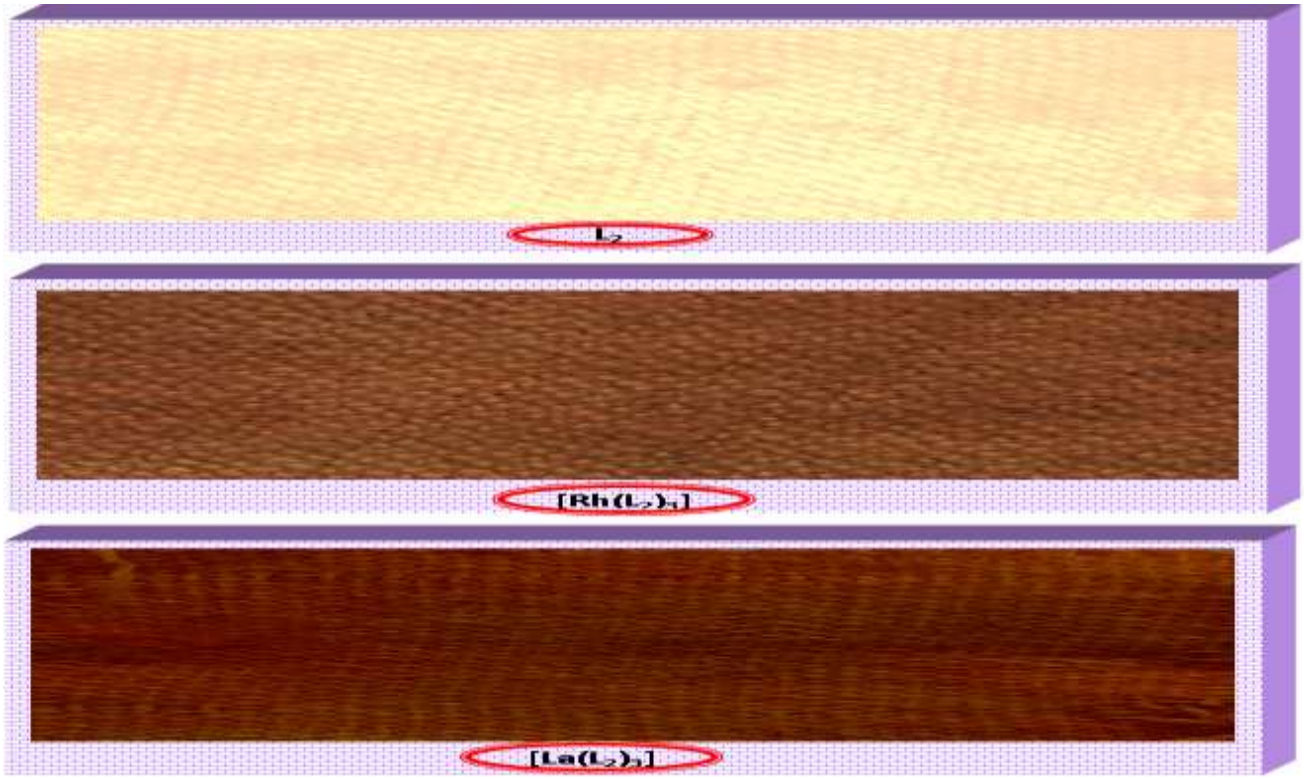


Fig.15: Samples the textiles dyeing of the ligand (HL_2) and their complexes

Conclusion

In this work, the metal ions complexes have been readied with the ligands. The willing compounds are described by melting point, flame atomic absorption, FT.IR and UV-Vis spectroscopy, as well conductivity quantifications. Exploration from antimicrobial activities was lifted out opposite the experimented organism. The dyes as well their produced compounds were applied at cotton fabric. According the result data an octahedral structure suggested for prepared complexes.

References

1. Anitha Reddy KRV, Fasiulla (2015) Studies on metal (II) complexes of bisazo dye 2,2-[benzene-1,3-diyl di(E) diazene 2,1-diyl] dianiline, Arch. Appl. Sci. Res.,7(1): 59-65.
2. Chaulia SN (2016) Metal complexes of multidentate azo dye ligand derived from 4-aminoantipyrine and 2, 4-dihydroxybenzoic acid; synthesis, characterization and biological activity, Der. Pharma. Chemica, 8(3): 254-272.
3. Weglarz-Tomczak E, Gorecki T (2012) Azo dyes-biological activity and synthetic strategy, Science, 66(12): 1303-1307.
4. Canakci D, Saribigik OY, Serin S (2014) Synthesis, structural characterization of Co (II), Ni (II) and Cu (II) complexes of azo dye ligands derived from dihydroxy naphthalene, Tnter. J. Sci. Res. Innov. Tech., 1: 52-72.
5. Otutu JO (2013) Synthesis and application of azo dyes derived from 2-amino-1, 3, 4-thiadiazole-2-thiol on polyester fibre, Inter. J. Res. Rev. Appl. Sci., 15: 292-296.
6. Chhowala TN, Desai KR (2015) Synthesis of Cu (II) and Ni (II) azo complex dyes, their application on silk fabries and screening for antibacterial activity, Inter. J. Sci. Res., 4: 901-905.
7. Hrdina R, Lustinec D, Stolin P, Burgrert L, Holdapek M (2004) Iron complexes of reactive azo dyes, Advance in Colour Science and Technology, 7(1): 6-17.
8. Valentina C, Sebez TI (2012) Azo dyes complexes, synthesis and tinctroial properties, U.P.B. Sci. Bul., 74: 109-118.
9. Dhahir SA, Aziz NM, Bakir SR (2012) Synthesis, characterization and antimicrobial studies of complexes of some metal ions with 2-[2-amino-5-(3,4,5-trimethoxy-benzyl)-pyrimidinyl-4-azo-4-bromo-phenol], Int. J. Basic and Appl. Sci., 12(6): 58-67.
10. Nair MLH, Mathew G, Kumar MRS (2005) Synthesis and characterization of some new Cu(II) complexes of azo dyes derived from 1,2-dihydro-1,5-dimethyl-2-phenyl-4-amino-3H-pyrazol-3-one, Indian. J. Chem., 44A: 85-89.
11. Andrews JM (2005) The BSAC working party on susceptibility testing of the British Society for antimicrobial chemotherapy, BSAC standardized disc susceptibility testing method (version 4), J. Antimicrobial. Chemothrap., 56(1): 60-76.

12. Raman N, Raja JD, Sakthivel A (2007) Synthesis, spectral characterization of Schiff base transition metal complexes: DNA cleavage and antimicrobial activity studies, J. Chem. Sci., 199(4):303-310.

13. Patel PS, Hathi MV (2010) Studies on synthesis and dyeing performance of disperse azo dyes based on Schiff base of ninhydrin and 4-amino phenol, J. Chem. Pharm. Res., 2(6):78-85.

14. Nair MLH, Sheela A (2008) Synthesis, spectral, thermal and electrochemical studies of oxomolybdenum (V) and dioxomolybdenum (VI) complexes of an azo dye derived from 4-amino-2, 3-dimethyl-1-phenyl pyrazole-5-one, Indian. J. Chem., 47A: 1787-1792.

15. Silverstein RM, Webster FX (1996) Spectrometric identification of organic compounds, John Wiley and Sons, 6th Ed, New York, 180.

16. Faghihi K, Hagibeygi M (2007) New aromatic polyamide with azo and phosphine oxide groups in the main chain, Turk. J. Chem., 31: 65-73.

17. Bartta P (2006) Sulpher bridged ruthenium-molybdenum complexes :(R-aapm) 2Ru (μ-S) $_2$ Mo (OH) $_2$ Synthesis, spectroscopic and electrochemical characterization. R-aapm = 2-(arylazo) pyrimidine, J. Chem. Sci., 118(2):171-177.

18. Pal S, Sinho C (2001) Studies on the reactivity of cis-$RuCl_2$ fragment in $Ru(pph_3)_2(TaiMe)Cl_2$ with N,N-chelators (Taime=1-Methyl-2-(p-Tolylazo) Imidazole). Spectral and electrochemical characterization of the products, Proc. Ind. Acad. Sci., 113 (3): 173-182.

19. Geary WJ (1971) Characterization of coordination compounds, Co ord. Chem. Rev., 7: 81-122.

20. Vadher GB, Zala RV (2011) Synthesis and analytical studies of some azo dyes as ligands and their metal chelates, Int. Chem. Sci., 9(1):87-94.

21. Nair MLH, Lalitha KP (2014) Synthesis, physicochemical studies and structure determination of some novel manganese (III) complexes of azo dyes derived from 4-aminoantipyrine, Int. J. Sci. Res. Publ., 4(2): 1-4.

22. Lever ABP, Soloman EI (2014) Inorganic electronic structure and spectroscopy, Wiley and Sons.

23. Al-Naimi SMH, Kindeel AS, Jarad AJ, Al-Noor TH (2016) Synthesis and antibacterial activities of some mixed ligand complexes, Trans. Engin. Sci., 4(2): 42-47.

24. Jarad AJ, Kadhim ZS (2015) Synthesis, spectral, dyeing performance and biological activity studies of azo dyes complexes with some metal ions, Int. J. Human. Arts. Med. Sci., 3(10): 197-210.

25. Ogunniran KO, Ajanaku KO, James OO, Ajani OO, Adekoya JA, Nwinyi OC (2008) Synthesis, characterization, antimicrobial activity and toxicology study of some metal complexes of mixed antibiotics, African. J. Pur. Appl. Chem., 2(7): 069-074.

26. Patel BK, Patel SD (2017) Studies on transition metal chelates of azo ligand containing bon acid, J. Phram. Sci. Biosci. Res., 7(1): 148-152.

27. Al-Khafagy AH (2016) Synthesis, characterization and biological study of some new metal-azo chelate complexes, J. Chem. Pharm. Res., 8(8): 296-302.

28. Pallikavil R, Umnathur MB, Krishnankuty K (2012) Schiff bases tetrephthaladehyde with 2-aminophenol and 2-aminothiophenol and their metal complexes, Arch. Appl. Sci. Res., 4: 223-2227.

29. Khalil MMH, Ismail EH, Mohamed GG, Zayed EM, Badr A (2012) Synthesis and characterization of novel Schiff base metal complexes and their application in determination of iron in different types of natural water, Open. J. In org. Chem., 2: 31-21.

30. Jarad AJ (2013) Synthesis and characterization of 6-(4-nitrobenzene azo)-3-amino benzoic acid complexes with Y (III) and La (III) ions, Eur. Chem. Bull., 2(6): 383-388.

