Curr. Pharm. Res. **2019**, *418,* 144-153

Current Pharma Research
ISSN-2230-7842
CODEN-CPRUE6
www.jcpronline.in/

Research Article

Theme- *New horizons in chemical sciences.*
Guest Editor- *R.P. Pawar*

Synthesis, Characterization and Biological Activity of Transition Metal Complexes of Schiff Base Ligand containing 1, 3, 4 Thiadiazole Moiety.

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Received 17 March 2019; received in revised form 15 September 2019; accepted 09 October 2019

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ABSTRACT

1,3,4 Thiadiazole moieties containing Ligand and their metal complex of Fe(III), Co(II), Ni(II) was prepared by using 5-amino-1,3,4-thiadiazole-2-thiol and substituted Salicyladehyde derivatives. The ligand and its metal complexes were analyzed and characterized by different spectroscopic techniques (UV, FT-IR, 1HNMR, 13CNMR, HRMS), Magnetic Susceptibility and molar conductance, elemental analysis. The transition metal complexes show moderate to excellent antifungal activity against Aspergillus Niger and Fusarium Oxysporum and antibacterial activity against Staphylococcus aureus and Bacillus subtilis using Kirby-Bauer disc diffusion method. The synthesis of new potential metal derived drug.

KEYWORDS

1, 3, 4 Thiadiazole, Metal Complexes, Magnetic Susceptibility, Antifungal activity, Antibacterial activity.

1. INTRODUCTION

The Most of the heterocyclic moieties having biological activity that depends on their Structure and their orientation[1], 3,4Thiadiazole is important compound because of their pharmaceutical, biological and analytical application[2]. 3, 4 thiadiazole also acts as ligands, increases biological activity by forming complex[3]. Schiff base is a condensation product of aldehyde with primary amine important in organic synthesis and other applications[4]. The aldehyde are ortho-substituted with hydroxyl group, which acts as a bidentate ligand for transition metal ions[5]. Schiff base is very important because of their structural similarity and flexibility with naturally occurring biological substance. Mine group >C=N- (azomethine) also helps to elucidate the racemization and transformation in biological systems[6]. during last few years more focus on different Thiadiazole derivative because of their important biological properties such as analgesic[7], anti-inflammatory[8], antituberculosis[9], antimicrobial[10], anticonvulsants[11], antihypertensive[12], antioxidant[13], anticancer[14], antifungal[15], antidepressant[16].

The present work is concerned with the Synthesis and Characterization and biological analysis of 1, 3, 4 Thiadiazole moieties containing ligand Scheme.1 and its Fe (III), CO (II) and NI (II) Metal Complexes Fig.1.

2. MATERIALS AND METHODS

2.1. Experimental

All the chemicals are of analytical grade. All salts are metal nitrates i.e. Fe $(NO_3)_3.9H_2O$,$CO(NO_3)_2$ $.6H_2O$,$Ni(NO_3)_2.6H_2O$ were purchased from Sigma-Aldrich and used without further purification. 5-amino-1,3,4-thiadiazole-2-thiol and 3,5-dichloro-2-hydroxybenzaldehyde from Sigma-Aldrich and used without further purification diethyl ether (Sigma-Aldrich), were purchased and used without purification. Distilled Ethanol was used for synthesis IR Spectra were recorded on Perkin Elmer Spectrometer in range 4000-400 cm-1 KBr pellets. 1H and 13CNMR Spectra were recorded in solvent DMSO using TMS as a Standard on BRUKER AVANCE III HD NMR 500 MHz spectrophotometer and chemical shift were recorded in ppm. Room Temperature magnetic moments by Guoy's method. Electronic Spectra was carried out using DMSO on Varian Carry 5000 Spectrometer. Molar Conductance measurements of metal complexes in dry DMSO having 1×10-3 concentration were carried out using Systronics Conductivity Bridge at room temperature. Elemental analysis (C, H and N) of the compound were carried out by using perkin Elmer 2400 elemental analyser. Mass Spectra were recorded on Bruker IMPACT HD.

2.2. Pharmacology

Schiff Base and their metal complexes evaluated in vitro their antibacterial activity against two Gram-Positive bacteria, viz, Staphylococcus aureus, Bacillus Subtilis; Two fungal strains Aspergillus niger and Fusarium oxysporum by Kirby-Bauer disc diffusion method[17]. The fungal and bacterial strains sub cultured on Potato dextrose agar and Nutrient Agar. The stock solution of (1 mg mL^{-1}) of the test chemical was prepared in DMSO solution. The stock solution again diluted by using sterilized water to different dilution in 500 ppm. The test chemicals of

different dilution were inoculated with help of sterilized micropipette on sterilized antimicrobial susceptibility discs. The bacteria were subculture in agar medium and disc were kept incubated for 36^{o}C at 24 hrs. The standard antibacterial drug Ciprofloxacin and Miconazole was also screen under same condition for comparison. Activity was measure by zone of inhibition (mm) surrounding discs. The experimental value compare with standard drug Ciprofloxacin for the antibacterial activity and Miconazole for the Antifungal activity.

2.3. Method for synthesis of Schiff base Ligand

The condensation of 1:1 5-amino-1,3,4-thiadiazole-2-thiol1 (1.33g,0.01mol) with 3,5-dichloro-2-hydroxybenzaldehyde 2(1.91g,0.01mol) dissolved in ethanol. The reaction mixture stirred for 30 min. at 60^{o}C. Few drops of glacial acetic acid were added to keep P^{H} 5. The resultant mixture stirred for 3-4 hrs., the colored precipitate of Schiff base ligand 2,4-dichloro-6-(((5-mercapto-1,3,4-thiadiazol-2-yl)imino)methyl)phenol 3, was obtained, which washed with Ethanol and finally recrystallized with Ethanol and Ether then dried in air. The purity of compound was checked by TLC using Silica Gel.

2.4. Method for synthesis of Metal Complexes

The metal complexes were prepared by mixing of ethanolic solution of $Fe(NO_3)_3.9H_2O$,$CO(NO_3)_2.6H_2O$,$Ni(NO_3)_2.6H_2O$ with (30ml) ethanolic solution of Schiff base (Fig. 1) in 1:2 (metal: ligand) ratio. The resulting mixture refluxed on water bath for 6-7hr. A colored product obtain filtered, washed with ethanol and recrystallized with ethanol. It was further dried in electric oven at 50-70^{o}C and recrystallized with Ethanol. Metal Complexes are bis(2,4-dichloro-6-(-((5-mercapto-1,3,4-thiadiazol-2-yl)imino)methyl)phenoxy) iron dihydrate, bis(2,4-dichloro-6-(-((5-mercapto-1,3,4-thiadiazol-2yl)imino)methyl)phenoxy)cobalt dihydrate,bis(2,4-dichloro-6-(-((5-mercapto-1,3,4-thiadiazol-2-yl)imino)methyl)phenoxy)nickel dihydrate (Fig.1).

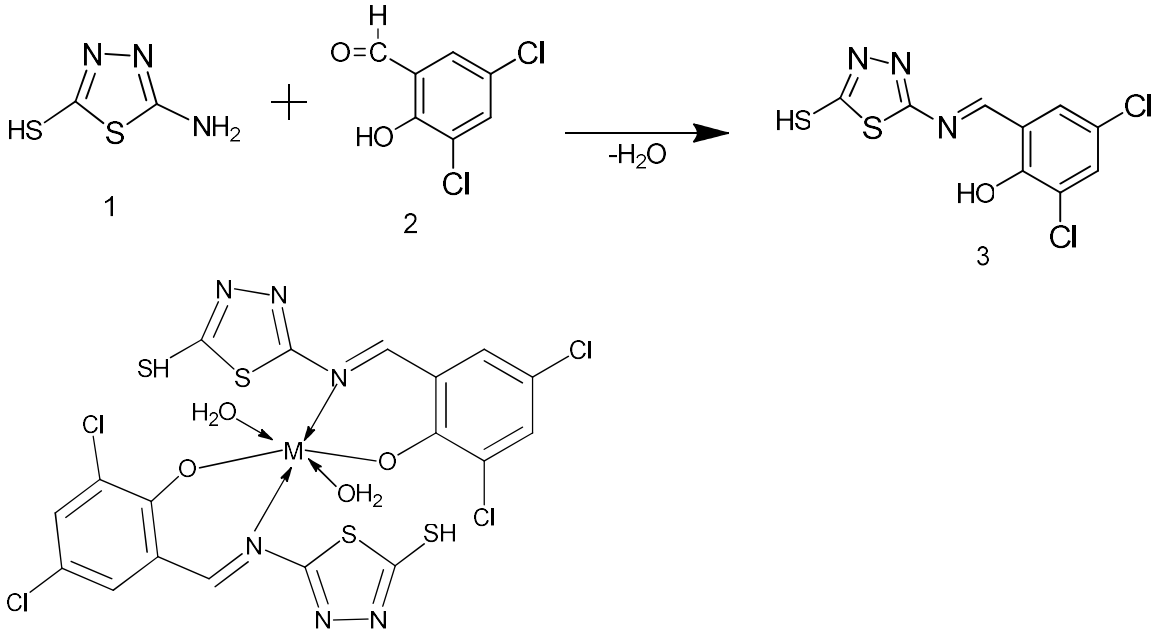

Fig.1. Proposed Structures of metal complexes M:Fe(III),CO(II) and Ni(II).

3. RESULTS AND DISCUSSION

The synthesized ligand (Scheme 1) and its transition metal complexes of 2,4-dichloro-6-(5-mercapto-1,3,4-thiadiazol-2-yl)imino methyl phenol Fig.1 are stable at room temperature in solid state. The ligand is soluble in organic solvent and metal complexes are easily soluble in DMSO. The synthesized complex having 1:2 metal to ligand stoichiometric ratio. The analytical and physical data shown in Table 1. Spectral data shows formation of ligand and its metal complexes.

3.1. IR Spectra

The IR data of the spectra of 2,4-dichloro-6-(5-mercapto-1,3,4-thiadiazol-2-yl)imino)methyl phenol (HL) Schiff base and its complexes are listed in Table 2. The IR spectra of the complexes are compared with those of the free ligand in order to determine the coordination sites that may be involved in a chelation. There are some guide peaks in the spectra of the ligand, which is different in metal complex helps to prove formation of metal complexes IR spectra of (2,4-dichloro-6-(5-mercapto-1,3,4-thiadiazol-2-yl)imino)methyl phenol (HL)Schiff base ligands exhibited the most characteristic bands at 3319-3330 cm-1ν(O-H), 1640-1650 cm-1 ν(C=N, azomethine) and 1260-1270 cm-1 ν(C-O). The ligand spectra showed bands at 3316-3304 and 1340-1348 cm–1 due to the stretching and deformation of the phenolic OH (Nakamoto et al. 1998). These are absent in the spectra of the complexes indicates the deprotonation of the hydroxyl group and co-ordination through phenolic oxygen. The band 1,640–1,648 cm-1 due to the azomethine group of the Schiff bases have shifted to lower frequency (1,612–1,633 cm^{-1}) after complexation, indicating the bonding of nitrogen of the azomethine group to the metal ion and this can be explained by the donation of electrons from the nitrogen to the empty d-orbital of the metal ion[18-19]. The phenolic λ(C–O) stretching vibration that appeared at 1,260–1265 cm^{-1} in Schiff bases shift towards higher frequencies (20–31 cm^{-1}) in the complexes. This shift confirms the participation of oxygen in the C–O–M bond. The appearance of broad bands around at (3,375–3,419 cm-1) in the spectra of complexes may be due to water molecules[20]. Two new bands appearing in the low frequency range 530–572 cm-1 and 464–470 cm-1 are assigned to ν(M–O) and ν(M–N), respectively. The ν(C–S–C) at 750–752 cm-1 of the Thiadiazole ring remain unchanged suggested that Thiadiazole group does not coordinate to the metal ion by neither nitrogen nor sulphur atom of Thiadiazole ring (Table 2) [21-22].

Table 1. Analytical Data and physical properties of ligand and its metal complexes.

Comp.	Empirical Formula	Mol.Wt.	Color	M.P (°C)	Yield (%)	Elemental Analysis/ Found (Calc.)			
						C	H	N	S
Ligand (HL)	$C_9H_5Cl_2N_3OS_2$	306	Dark Yellow	118	72	35.89 (35.30)	1.69 (1.65)	13.63 (13.72)	20.21 (20.94)
Fe (III) Complex	$C_{18}H_{12}Cl_4FeN_6O_4S_4$	702	Brown	>300	71	29.30 (30.79)	1.81 (1.72)	11.87 (11.97)	19.15 (18.26)

Co(II) Complex	$C_{18}H_{12}Cl_4CoN_6O_4S_4$	705	Light red	>300	73	30.96 (30.65)	1.64 (1.71)	11.25 (11.92)	18.98 (18.18)
Ni(II) Complex	$C_{18}H_{12}Cl_4NiN_6O_4S_4$	705	Green	>300	71	29.68 (30.66)	1.23 (1.72)	11.45 (11.92)	18.87 (18.19)

3.2. ^{1}H NMR and ^{13}C NMR Spectra

The 1H-NMR spectra of ligand were recorded in DMSO-solution using TMS as a standard. The spectra of ligands shows singlet at δ 7.19-7.89 ppm due to aromatic proton while azomethine proton resonate at singlet δ 8.9 ppm the phenolic OH has signal singlet at δ 11.20 ppm and Thiadiazole containing –SH group shows singlet at δ 13.43 ppm[23].^{13}C NMR of Ligand, peak appeared at δ158-162 ppm imine group(-C=N-),peak 187.50ppm Due to C-SH bonding in Thiadiazole.121.96-135.53 ppm due to aromatic carbon,158-170 ppm peak due to Ar-OH group(Table 3)[24].

Table 2. Infrared Spectra of the Schiff base and Complexes in Cm^{-1}.

Compound	**νOH/H_2O**	**νC-O**	**νC=N**	**νM-N**	**νM-O**	**νC-S-C**	**ν-C=N-N=C**	**νN-N**
Ligand	3319	1265	1645	___	___	752	1467	1028
Fe(III) Complex	3388	1307	1633	468	530	754	1433	1022
Co(II) Complex	3375	1280	1622	470	574	755	1432	1028
Ni(II) Complex	3419	1282	1612	464	572	755	1485	1028

Table 3. ^{1}H NMR Signals (δ, ppm) and their assignments.

Compound	**^{1}H NMR Signals (δ,ppm) and their assignments**
Ligand(HL)	11.20 (s,1H,Ar-OH), 8.9(s,1H,CH=N),7.19-7.89 (s,2H,Ar-CH), 13.43 (s,1H,SH)

3.3. Mass Spectra

Mass Spectra of ligands shows peak at m/z 305 which is M+H peak at 100% intensity this peak support to the structure of the ligand.

3.3.1. Magnetic Susceptibility and molar conductance

The magnetic susceptibility observed at room temperature. The entire synthesized metal complex is paramagnetic in nature. Molar conductance values of metal complexes were observed at room temperature at 1×10^{-3}M DMSO Solution. The studies show negligible molar conductance value in range 8-12 ohm^{-1} cm^2 mol^{-1} results shows in table 4. It is clear that all metal complexes are non-electrolytic in nature[25-27].

3.3.2. Electronic absorption Spectra

The electronic spectral data of the ligands and metal complexes in DMSO solution are given in Table 4. The nature and geometry of the ligand field around the metal ion has been deduced from the electronic spectra data. The band appearing at 220-310 is due to transition of benzene ring of the ligand. The other band due to free ligands 320-380 nm due to transition for phenolic OH and azomethine moieties. These band shifts longer due to formation of Schiff base metal complexes[26]. The spectra of the complexes display band 424-500 nm assigned to charge transfer transition from ligands to metal[27]. The magnetic moment value for CO(II) complexes is 5.08 B.M is near to octahedral complex spectra shows two band at 480-500 nm and 702-710 nm shows that octahedral geometry of CO(II) complexes[28-29].Electronic spectra of Ni(II) complexes shows three band 950 nm,549 nm and 408 nm suggest octahedral geometry[30-32].

Table 4. Electronic spectral Magnetic and Molar conductance Data.

Compounds	**Wavelength in nm**	**Magnetic moment μeff (BM)**	**Molar conductance (ohm-1cm2 mol-1)**
Ligands (HL)	260,370	--	6.68
$C_{18}H_{12}Cl_4FeN_6O_4S_4$	396-401,460-456,591-596	5.92	12
$C_{18}H_{12}Cl_4CoN_6O_4S_4$	310-350,480-500,703-710	5.08	9.5
$C_{18}H_{12}Cl_4N_6NiO_4S_4$	376-408,549,950	3.12	11.09

3.4. Antimicrobial activity

The in vitro Antimicrobial activity of the synthesized ligand and their corresponding metal complexes on selected two gram positive *bacteria S. aureus and B. Subtlis* two fungi *A. niger* and *F. Oxysporum* was carried out (Table 5). All of the tested compounds showed good biological activity against test microorganism. The bactericidal and fungicidal investigation data of the compounds are summarized in Table 5. The investigation showed that CO(II) And Ni(II) shows more, bactericidal and fungicidal activity than Fe(II) Complexes and Ligand hence activity of metal complexes increases due to chelation increase in delocalization of π electron on chelating ring and enhance the penetration of complexes in lipid membrane and blocks the binding site enzymes of microorganism there are other factors also, solubility, lipophilicity/hydrophilicity, Conductivity and M-L bond length that increases the activity of complexes[33-35].

Table 5. Antimicrobial activity of ligand and its Metal Complexes.

Comd.	Antibacterial Activity				Antifungal Activity			
	S. aureus		***B. subtilis***		***A. niger***		***F. oxysporum***	
	Diameter of inhibition Zone in mm	% Activity Index	Diameter of inhibition Zone in mm	% Activity Index	Diameter of inhibition Zone in mm	% Activity Index	Diameter of inhibition Zone in mm	% Activity Index
	ppm							
	500	500	500	500	500	500	500	500
Ligands (HL)	22	65	21	64	20	65	18	67
Fe	19	56	16	48	19	61	14	52
Co	26	76	22	67	25	81	24	89
Ni	24	71	23	70	23	74	20	74
Ciprofloxacin (Standard)	34	100	33	100	--	--	---	--
Miconazole (Standard)	--	--	--		31	100	27	100

4. CONCLUSION

In the present work our efforts to synthesize and characterize some novel compounds from conventional methods. These compounds characterized by physicochemical and spectral analyses. The synthesized Schiff base ligand binds metal ions in bidentate manner, with NO donor site of azomethine-N and deprotonated phenolic-O.the antimicrobial activity data showed that Most of the metal complexes are more biologically active compared to those parent ligand against all pathogenic species. Such studies may help to decrease emerging problems in drug resistance in health sciences.

5. ACKNOWLEDGEMENTS.

We are thankful to the Head of C.I.F S.P.P.U Pune**,** S.A.I.F Panjab University, Chandigarh, J.E.S College Jalna, Shivaji Arts, Commerce and Science college Kannad, Aurangabad and Pratishthan Mahavidyalaya, Paithan, Aurangabad India For their support.

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