69

A new series of Schiff bases derived from o- vanillin and dapsone: Synthesis, spectral, thermal and antibacterial studies

¹V. A. Sadafale, ²P. R. Mandlik

¹Assistant Professor, ²Professor Department of Chemistry, Adarsha Science, J. B. Arts & Birla Commerce Mahavidyalaya, Dhamangaon Rly, Amravati, India

Abstract: A novel Schiff base ligand was prepared by the condensation of o- vanillin and dapsone, and then metal complexes of Co(II), Ni(II), Cr(III) and VO(IV) were synthesized. The metal complexes were characterized and investigated by physical and spectral techniques, namely, elemental analysis, melting point, conductivity, 1H NMR, IR, UV-Vis spectra, ESR, and mass spectrometry measurements. They were further analyzed by thermal technique (TGA/DTA) to gain better insight about the thermal stability and kinetic properties of the complexes. Thermal data revealed high thermal stability and non-spontaneous nature of the decomposition steps. The Freeman-Carroll and Sharp-Wentworth method was applied to extract thermodynamic parameters to explain the kinetic behavior. The molar conductance values were relatively low, showing their non-electrolytic nature. The electronic and ESR spectra of metal complexes provides information about geometry of the complexes and are in good agreement with proposed square planar geometry of Co(II) and Ni(II), octahedral for Cr(III) and square pyramidal for VO(IV) complexes. The antibacterial activity of all the compounds was screened against four bacterial pathogens, namely, E. coli, S. aureus, P. aeruginosa and K. pneumoniae. It has been found that the Schiff base and its metal complexes showed significant antimicrobial activity.

Index Terms: Dapsone, Schiff base, Metal complexes, Antibacterial activity

I. INTRODUCTION

The sulfonamides (SO₂–NH-) are very important protected intermediates of amines with several types of biological activities such as anti-bacterial, hypoglycemic, diuretic, anti-carbonic anhydrase (CA), antithyroid, anti-inflammatory, anticancer, anti-hypertensive, anti-convulsent and herbicidal for potential agricultural applications [1-4]. Among the pharmacological properties, antibacterial is the most important one. Sulfonamides inhibit the biosynthesis of tetrahydrofolate in the bacteria, which is a co-factor for the bacterial DNA and RNA synthesis. Deficiency of tetrahydrofolate, decreases the production of new DNA and RNA, ultimately resulting in the bacterial decay. On the other hand, many organic drugs, which possess modified pharmacological and toxicological properties administered in the form of metallic complexes, have the potential to act as ligands and the resulting metal–drug complexes are particularly important both in coordination chemistry and biochemistry , however, the study of metal–drug complexes is still in its early stages, thus representing a great challenge in current synthetic chemistry, coordination chemistry and medicinal bioinorganic chemistry . Sulfa drugs have attracted special attention for their therapeutic importance as they were used against a wide spectrum of bacterial ailments [5].

Recent advances in chelation research have concrete the way for targeting "metal complexes" for chemotherapy, using different strategies and pharmacological manipulation, demonstrating significant prospects for the utilization of metal complexes as drugs and presenting a thriving arena for medical inorganic biochemistry. The pharmaceutical use of metal complexes therefore has excellent potential [6], but comparatively less study is carried out.

In view of the above facts the present investigation is focused on synthesizing Sulfa drug Schiff base ligand and their complexes, finding out binding mode of ligands towards different metal ions and also studying the stereochemistry of complexes.

II. EXPERIMENTAL –

Reagents and materials-

All the metal salts and solvents used were of analytical reagent grade purchased from SD Fine and Merck. Pure sample of dapsone was obtained from Shah Scientific Pvt. Ltd. Mumbai. Solvents were purified and dried before use by literature method [7]. All the metal salts are commercially available and were used as received. The ligand used in the present work is not commercially available; hence, it was synthesized in the laboratory.

Physical measurements-

Microanalyses of carbon, hydrogen and nitrogen of the ligand were carried out on a Perkin Elmer CHN 2400 elemental analyzer The amounts of metal present in the complexes were estimated by metal oxide method [8]. The molar conductance of the complexes in dimethyl formamide (DMF) solution (10^{-3} M) , was measured using Equiptronic digital conductivity meter EQ-660 with a cell constant of 1.00 cm^{-1} at room temperature. The magnetic susceptibility of the complexes were measured by the Gouy method at room temperature using Hg[Co(NCS)₄] as a calibrant. ¹H-NMR spectrum of the ligand was recorded in DMSO-d₆ solution on EM-360, 60 MHz NMR Spectrometer. The electronic spectra of the ligand and complexes were recorded on a shimadzu UV/Vis spectrophotometer in the region 200-1000 nm. The infrared Fourier-transform infrared (IR) spectra of ligand and complexes were

recorded as KBr pellets using Shimadzu spectrometer (IRAffinity-1S). Mass spectra of synthesized compounds were carried out on Thermo scientific TSQ 8000 Gas Chromatograph- Mass Spectrometer. The thermal analysis was performed with a Perkin Elmer (TGS-2 model) thermal analyzer at a heating rate of 10° C min⁻¹ in the temperature range 40-500°C. ESR spectra of complexes at room temperature and liquid nitrogen temperature were carried by using JES-FA 200 ESR spectrometer. **Synthesis of ligand (L¹H)** -

Equimolar solutions of o- vanillin (1.58 g, 10 mmol) and dapsone (2.48g, 10 mmol) were mixed separately in ethanol (50 ml) and refluxed for 1 h. The solution was concentrated and cooled. Orange coloured precipitate was obtained which was filtered off and recrystallized from ethanol. The purity of the compounds were checked by thin layer chromatography (TLC). Yield 73%, M. P. 248^oC. It was characterized by elemental analysis, IR, UV, ¹H NMR and mass spectra.

¹H NMR δ 12.65 ppm (2H, S, -OH), δ 8.93 ppm (1H, S, azomethine, -HC=N), δ 5.90- δ 7.89 ppm (11H, M, Aromatic proton), δ 3.84 ppm (3H, S, -OCH₃) [9]. The schematic representation of synthesis of L¹H is shown in reaction scheme 1.



Scheme1. Synthesis of L¹H ligand

Synthesis of complexes-

The Schiff base was taken in ethanol (50 ml) and stirred gently and then equimolar ethanolic solution of $Co(OAc)_2.4H_2O$, $Ni(OAc)_2.4H_2O$, $CrCl_3.6H_2O$ or $VOSO_4.5H_2O$. Few drops of 1N KOH were added to each reaction mixture and were refluxed in water bath for about 10-12 h. The colored solids obtained were filtered off, washed several times with ethanol followed by petroleum ether and finally dried over fused calcium chloride.

III. RESULTS AND DISCUSSION -

In the present study, all the complexes are found to be colored solids, air stable and non-hygroscopic. The analytical data indicate 1:1 metal ligand stoichiometry. The complexes are insoluble in water and common organic solvents but soluble in coordinating solvents like DMF and DMSO. Lower values of molar conductance indicate that complexes are nonelectrolytes. The analytical and physical data of the ligand and its complexes are given in Table 1.

Compound	Eanmula wt	Colour	Found (Calc.) %					
Compound	rormula wt.		С	Н	Ν	Μ		
C20H18N2O4S	382.10	Orange	62.81(62.66)	4.74 (4.79)	7.33 (7.26)	-		
[Co(L ¹)]	535.43	Pale Brown	49.35 (49.51)	4.52 (4.63)	5.23 (5.37)	11.01 (11.15)		
[Ni(L ¹)]	535.49	Brown	49.37 (49.52)	4.52 (4.68)	5.23 (5.37)	10.97 (11.12)		
$[Cr(L^1)]$	558.37	Brown	43.02 (43.24)	4.15 (4.28)	5.02 (5.22)	9.31 (9.42)		
[VO(L ¹) ₂]	829.79	Brown	57.90 (58.06)	4.13 (4.27)	6.75 (6.85)	6.14 (6.25)		

Table 1.Elemental analysis, colour and formula weight of the compound.

Infrared spectra

The IR spectra provided valuable information regarding the nature of functional group attached to the metal ion. A medium broad band at 3089 cm⁻¹ due to intramolecular hydrogen bonded OH group in the spectrum of free ligand found to be lacking in the spectra of all the complexes indicating the deprotonation of the replaceable hydrogen atom during coordination [10-11]. The absorption peak for v(C=N) at 1595 cm⁻¹ in the spectrum of ligand is shifted to a lower frequency by 10-30 cm⁻¹ in all complexes due to coordination of azomethine nitrogen [12-13]. Further conclusive evidence of the coordination of the ligand with the metal ions via oxygen and nitrogen was shown by the appearance of new bands in the spectra of all the complexes at 429-442 cm⁻¹ and 550-565 cm⁻¹ which were assigned to the metal- nitrogen (M–N) and metal-oxygen (M–O) stretching vibrations, respectively [14]. A pair of bands corresponding to v(NH₂) at 3365-3379 cm⁻¹ and 3225-3242 cm⁻¹ were present in the spectra of ligand as well as metal complexes indicating non-involvement of the group in coordination with metal ion[15]. Two prominent bands, one at 1253-1294 cm⁻¹ and other at 1107-1134 cm⁻¹ are observed, which are due to the symmetric and asymmetric vibration of S-O bond of the sulphone group [16]. The spectra of Co(II), Ni(II) and Cr(III) complexes display four additional bands in the region 3440-3452 cm⁻¹, $\nu_v(OH)$, 1423-1450 cm⁻¹, $\delta(H_2O)$, 833-842 cm⁻¹, $\rho_r(H_2O)$ and 707-712 cm⁻¹, $\rho_w(H_2O)$, suggesting the presence of coordinated water in each of them [17]. An additional band at 977 cm⁻¹ in the spectrum of VO(IV) complex is assigned to v(V=O) mode of vibration [18]. The IR spectral data of the compounds are listed in Table 2.

70

Compound	v(OH) (Hydrogen	v(NH2)	v(C=N)	v(SO ₂)		v(M-O)	v(M-N)	v(H2O)	
	bonded)			asym	sym				
L ¹ H	3089	3325, 3236	1595	1253	1107	-	-	-	
[Co(L ¹)]	-	3373, 3225	1587	1294	1134	565	438	3440, 1438, 833, 711	
$[Ni(L^1)]$	-	3379, 3238	1564	1261	1112	550	442	3452, 1423, 842, 712	
$[Cr(L^1)]$	-	3365, 3228	1570	1290	1124	565	429	3444, 1450, 839, 707	
$[VO(L^1)_2]$	-	3371, 3242	1588	1292	1115	557	437	-	

Table 2. IR frequencies of ligand and its metal complexes (cm⁻¹).

Mass Spectra:

The FAB mass spectrum of the ligand shows molecular ion peak $[M]^+$ at m/z 382.10; the other important peaks appearing at m/z 248, 227, 221, 157, 151 and 93 correspond to $[C_{12}H_{12}N_2O_2S]^+$, $[C_{14}H_{13}NO_2]^+$, $[C_{14}H_{13}NO_3]^+$, $[C_6H_7NO_2S]^+$, $[C_8H_9NO_2]^+$ and $[C_6H_7N]^+$ fragment ions. The mass spectra of complexes show molecular ion peaks in good agreement with the empirical formula suggested by elemental analyses. The representative fragmentation pattern for the ligand is given in Scheme 2.



Electronic spectra and magnetic moment

The electronic absorption spectra of metal complexes were recorded in DMSO. UV spectrum of Co(II) complex exhibit absorption bands at 427 nmand 610 nm, which may be assigned to ${}^{2}A_{1g} \rightarrow {}^{2}B_{1g}$ and ${}^{2}A_{1g} \rightarrow {}^{2}E_{g}$ transition respectively. These bands clearly suggest square planar geometry for Co(II) complex [19]. Electronic spectra of Ni(II) complex display bands at 620 and 748 nm which are assigned to transition ${}^{1}A_{1g} \rightarrow {}^{1}A_{2g}$ and ${}^{2}A_{1g} \rightarrow {}^{2}B_{1g}$ respectively. These are consistent with square planar geometry for Ni(II) complex. Electronic spectra of Cr(III) complex displaying three bands around 296, 448 and 620 nm which are assigned to transition ${}^{4}A_{2g} \rightarrow {}^{4}T_{1g}(P)$, ${}^{4}A_{2g} \rightarrow {}^{4}T_{1g}(F)$ and ${}^{4}A_{2g} \rightarrow {}^{4}T_{2g}(F)$ transition respectively are consistent with octahedral geometry for Cr(III) complex [20]. Electronic spectra of VO(IV) complexes display four bands. First band 470 nm assigned to ${}^{2}B_{2} \rightarrow {}^{2}A_{1g}$ transition while second band at 829 nm assigned to ${}^{2}B_{2} \rightarrow {}^{2}E$ transition. Third and fourth band at 223 nm and 258 nm respectively attributed to intra ligand charge transition suggested square pyramidal geometry for the complex [21]. The magnetic moments, absorption region, band assignment and proposed geometry of the complexes are given in Table 3.

Complex	Absorption region (nm) Band assignment		µ _{eff} B. M.	Λ_{M} Ω^{-1} cm ² mol ⁻¹	Geometry
[Co(L ¹)]	427	${}^{2}A_{1g} \rightarrow {}^{2}B_{1g}$ ${}^{2}A_{1g} \rightarrow {}^{2}E_{g}$	2.12	8.81	Square planer
[Ni(L ¹)]	620 748		Dia.	10.81	Square planer
[Cr(L ¹)]	296 448 620		3.85	9.26	Octahedral
[VO(L ¹) ₂]	223 258 470 829	$\frac{\text{INCT}}{\text{INCT}}$ $\frac{^{2}\text{B}_{2g} \rightarrow ^{1}\text{A}_{1g}}{^{2}\text{B}_{2} \rightarrow ^{1}\text{E}}$	1.35	9.68	Square pyramidal

Table 3. Electronic spectral data of the complexes.

Thermal analyses

The thermograms of all the complexes show a gradual weight loss indicating decomposition by fragmentation with increase in temperature. Analysis of thermograms of these complexes shows that Co(II), Ni(II) and Cr(III) complexes decomposed in four stages while ligand and the VO(IV) complex in single stage. The TG curves of Co(II), Ni(II) and Cr(III), complexes were stable upto 60°C. Above 60°C there occurs a loss of one lattice water molecule from Co(II), Ni(II) and Cr(III), complexes [% wt loss, found/calc: Co(II): 3.42/3.36, Ni(II): 3.43/3.36, Cr(III): 3.27/3.22]. Further weight loss in Cr(III), Co(II) and Ni(II) complexes upto 240°C corresponds to removal of two coordinate water molecules from Cr(III) complex and one coordinate water molecule each from Co(II) and Ni(II) complex. [% wt. loss, found/cal: Cr(III): 6.49/6.44, Co(II): 3.42/3.36, Ni(II): 3.43/3.36]. In VO(IV) complex no weight loss observed upto 220°C indicates absence of lattice and coordinate water molecule. The presence of water molecule in the complexes is also supported by their IR spectra. In all the TG curves of complexes, continuous and rapid weight loss has been observed above 240°C corresponding to thermal degradation of free part of the coordinated ligand along with other groups present therein. A gradual weight loss above 340°C corresponds to the degradation of actual coordination part of the ligand. These weight losses are accompanied by DTA peaks at Co(II)- 70°C, 310°C, 420°C, 420°C, Ni(II)- 110°C, 150°C, 330°C, 470°C; Cr(III)- 350°C, 390°C, 460°C and VO(IV)- 120°C, 200°C, 210°C, 250°C.

Finally the horizontal part beyond 345^{0} C suggests formation of final decomposition product corresponding to respective metal oxides CoO, NiO, Cr₂O₃,V₂O₃. The thermal decomposition data of the compounds is given in Table 4 and thermograms of ligand L¹H and its complexes is given Fig. 1-5. The relative thermal stability on the basis of half decomposition temperature is found to be Co(II) = Cr(III) > Ni(II) > VO(IV) > L¹H

Table 4. The thermal decomposition data of the compounds.									
Complex	Half decom. Temp. (⁰ C)	Sten	Ea (k/J)		(n)	ΔS	ΔF	S*	
		Step	FC*	SW**	(11)	(J/mol/K)	(kJ/mol)	(kJ)	
L ¹ H	260 ⁰ C	Ι	21.75	21.38	0.93	-294.17	113.82	-39.17	
$[Co(L^1)]$	460 ⁰ C	IV	17.87	17.90	0.98	-278.71	105.11	-38.78	
$[Ni(L^1)]$	440 ⁰ C	IV	6.02	7.45	0.96	-279.07	95.37	-37.84	
$[Cr(L^1)]$	460°C	IV	13.94	13.71	0.96	-268.59	97.26	-38.88	
$[VO(L^1)_2]$	350°C	Ι	21.66	21.32	0.98	-247.40	99.10	-39.26	



1. $L^{1}H$





Figure 1-5. Thermograms (TGA/DTA) of ligand L¹H and its complexes

ESR spectra

The X band ESR spectra of VO(IV)complex recorded in the solid state at room temperature (RT) and liquid nitrogen temperature (LNT) and their parameters g_{\parallel} , g_{\perp} , A_{\parallel} and A_{\perp} have been calculated. Both spectra of VO(IV) complex show single line pattern, which indicates that single vanadium is present in the molecule i.e. monomeric complex. The observed spectral parameter for VO(IV) complex are: $g_{\perp} = 2.0138$, $g_{\parallel} = 1.9618$ suggesting that the resulting complex is covalent and show the characteristics of square pyramidal geometry. The mononuclear vanadium complex show well resolved axial anisotropy with $g_{\parallel} < g_{\perp}$ a relationship characteristic of an axially compressed (dxy)¹ configuration. This indicates that the unpaired electron is present in d_{xy} orbital [22].

The ESR spectral parameters and typical ESR spectra of VO(IV) complex at room temperature and liquid nitrogen temperature are listed in Table 5 and shown in fig. 6 and 7.

Table 5.g and A	value of powder ESI	R spectra of VO(IV) complex of ligand L	¹ H at RT and LNT.

Complexes		Spectral Parameters								
		g∥	g⊥	/g/	A	A⊥	/A/			
VO(IV)	RT	1.9269	2.0052	1.9791						
	LNT	1.9618	2.0138	1.9964						



On the basis of the above characterization, the proposed structures of complexes are shown in Fig. 8.



M = Co(II), Ni(II)

M = Cr(III)

75



Figure 8. Suggested structures of metal complexes of ligand L¹H

IV. ANTIBACTERIAL ACTIVITY

All the compounds were screened for their antibacterial activity. The growth of *E. coli* was more inhibited by almost all the compounds as compared to other bacterial species. Ligand (L_1H) is highly active against *K. pneumoniae* whereas Cr(III) complex were found to show good activity against *E. coli*. All the bacterial pathogens tested were found to be resistant to Co (II) complex. The Ni(II) and VO(IV) complexes showed weak or good activity against all bacteria. Results of studies of antibacterial effect of ligand and its all metal complexes are summarized in Table 6.

S.N.	Ligand and its complexes	E. coli (mm)	S. aureus (mm)	P. pneumoniae (mm)	K. Pneumoniae (mm)
1	$L^{1}H$	S ₂₃	S ₂₅	S_{21}	S ₃₂
2	Co- L ¹ H	S ₁₂	\mathbf{S}_{12}	\mathbf{S}_{13}	\mathbf{S}_{12}
3	Ni- L ¹ H	S ₁₆	S ₁₇	\mathbf{S}_{12}	S_{18}
4	Cr- L ¹ H	S ₁₅	\mathbf{S}_{14}	\mathbf{S}_{11}	S ₁₃
5	VO-L ¹ H	S ₁₀	R	R	R

Table 6- Antibacterial activity of ligand and its complexes.

S- Sensitive (Bactericidal)

R-Resistant (Bacteriostatic)

CONCLUSION

The present article includes the synthesis and characterization of new Schiff base ligand and its Co(II), Ni(II), Cr(III) and VO(IV) complexes. Characterization of compounds includes IR, mass, ¹H NMR, ESR and TGA. Electronic spectrum suggests square planar geometry for Co(II) and Ni(II) complexes, octahedral geometry for Cr(III) and square pyramidal geometry for VO(IV) complex. The thermal study revealed that complexes are thermally stable. The relative thermal stabilities of the complexes was in the order Co(II) = Cr(III) >Ni(II) >VO(IV) > L¹H. ESR spectra of VO(IV) complex indicates that the unpaired electron is present in d_{xy} orbital The Schiff base ligand and its complexes have been tested in vitro to evaluate their antimicrobial activity against *Escherichia coli, Staphylococcus aureus, Pseudomonas aeruginosa* and *Klebsiella pneumoniae* using well-diffusion method. It has been found that the Schiff base ligand and its complexes show significant antimicrobial activity.

V. ACKNOWLEDGMENT

The authors are thankful to Principal, Jagadamba Mahavidyalaya, Achalpur, for providing necessary laboratory facilities, Principal, Adarsha Mahavidyalaya, Dhamangaon Rly. and Head, Department of Microbiology, Adarsha Mahavidyalaya, Dhamangaon Rly., for antimicrobial activity, Director RSIC, Chandigarh, for recording ¹H NMR spectra and mass spectra and Director CDRI, Lucknow, for elemental analysis. The authors express sincere thanks to Director IIT, Powai, for recording ESR spectra.

REFERENCES

[1] A. J. McCarroll, T. D. Bradshaw, A. D. Westwell, C. S. Matthews, M. F. G. Stevens, "Quinols as novel therapeutic agents. 7.1 Synthesis of antitumor 4-[1-(arylsulfonyl-1H-indol-2-yl)]-4-hydroxycyclohexa2,5-dien-1-ones by Sonogashira reactions," J. Med. Chem., 50 (7), 1707-1710, 2007.

[2] A. A. Kadi, N. R. El-Brollosy, O. A. Al-Deeb, E. E. Habib, T. M. Ibrahim, A. A. El-Emam, "Synthesis, Antimicrobial, and Antiinflammatory Activities of Novel 2-(1-Adamantyl)-5-substituted1,3,4-oxadiazoles and 2-(1-Adamantylamino)-5-substituted-1,3,4- thiadiazoles," Eur. J. Med. Chem., 42, 235-242, 2007.

[3] S. Schenone, C. Brullo, O. Bruno, F. Bondavalli, A. Ranise, W. Filippelli, B. Rinaldi, A. Capuano, G. Falcone, "New 1,3,4-thiadiazole derivatives endowed with analgesic and antiinflammatory activities," Bioorg. Med. Chem., 14(6), 1698-1705, 2006.

[4] M. M. Ghorab, F. A. Ragab, H. I. Heiba, R. M. El-Hazek, "Anticancer and radio-sensitizing evaluation of some new thiazolopyrane and thiazolopyranopyrimidine derivatives bearing a sulfonamide moiety," Eur. J. Med. Chem., 46, 5120-5126, 2011.

[5] A. I. Fatima Al-Khodir, "Ca(II), Zn(II) and Au(III) Sulfamethoxazole Sulfa-drug Complexes: Synthesis, Spectroscopic and Anticancer Evaluation studies," Orient. J. Chem., 31(3), 1277-1285, 2015.

[6] S. S. Ahmad Shah, M. Ashfaq, T. Najam, M. M. Ahmed, S. Shaheen, R. Tabassum and S. A. Ejaz, "Synthesis of Sulfonamides, Metal Complexes and the Study of In vitro Biological Activities," Current Bioactive Compounds, 9 (3), 2013.

[7] B. S. Furniss, A. J. Hannaford, P. W. G. Smith, A. R. Tatchell, Vogel's Textbook of Practical Organic Chemistry, 5th Ed.,Longmans, London 1989.

[8] A. S. Aswar, P. J. Bahad, A. V. Paradhi and N. S. Bhave, "Structural, semiconducting and thermal studies of some Schiff base coordination polymers J. Polym. Mater., 5, 232,1988.

[9] M. B. Halli and V. B. Patil, "Synthesis, spectral characterization and DNA cleavage studies of Co(II), Ni(II), Cu(II), Zn(II), Cd(II) and Hg(II) complexes with benzofuran-2-carbohydrazide schiff bases", Indian J. Chem., 50A, 664-669, 2011.

[10] M. M. Abd- Elzaher, A. A. Labib, H. A. Mousa, S. A. Moustafa, M. M. Ali and A. A. El- Rashedy, "Synthesis, anticancer activity and molecular docking study of Schiff base complexes containing thiazole moiety," Beni-Suef University of Basic and Applied Sciences, 5, 85-96, 2010.

[11] D. Gurbuz, A. Cinaril, A. Tavman and S. B. Tan, "Synthesis, characterization and antimicrobial activity of some transition metal complexes of n-(5-chloro-2-hydroxyphenyl)-3- methoxy-salicylaldimine," Bull. Chem. Soc. Ethiop., 29(1), 63-74, 2015.

[12] A. A. Kachare, S. A. Kumbalpuri, S. G. Shankarwar and T. K. Chondhekar, "Synthesis, characterization and biological studies of some transition metal complexes of hydrazone Schiff base ligand derived from of 2-quinolone and 1-phthalazine hydrazine hydrochloride," Der ChemicaSinica, 6 (7), 87-92, 2015.

[13] O. B. Ibrahim, M. A. Mohamed and M. S. Refat, "Nano Sized Schiff Base Complexes with Mn(II), Co(II), Cu(II), Ni(II) and Zn(II) Metals : Synthesis, Spectroscopic and Medicinal Studies," Canadian Chemical Transactions, 2(1), 108-121, 2014.

[14] M. Asadi and M. S. Khah, "Some new unsymmetrical diiminotetradentateschiff base derived from 3,4-Diaminobenzophenone: Synthesis, characterization and the formation constant of Ni(II) and Cu(II) complexes J. Iran. Chem. Soc., 7(4), 875, 2010.

[15] N. I. Taha, "Synthesis of 1,3-Oxazepine Derivatives Derived from 2-(1H-Benzo[d][1,2,3]Triazol-1-yl) Acetohydrazide by Using Microwave Irradiation", International Journal of Organic Chemistry, 7(3), 219-228, 2017.

[16] A. Mumtaz, T. Mahmud, M. R. Elsegood and G. W. Weaver, "Synthesis and Characterization of New Schiff Base Transition Metal Complexes Derived from Drug Together with Biological Potential Study," J. Nucl. Med. Radiat. Ther. 7(6), 310-315, 2016.

[17] A. S. Munde, A. N. Jagdale, S. M. Jadhav and T. K. Chondhekar, "Synthesis, characterization and thermal study of transition metal complexes of an asymmetrical tetradentate Schiff base ligand", J. Serb. Chem. Soc., 75(3), 344, 2010.

[18] S. Annapoorani and C. N. Krishnan, "Synthesis And Spectroscopic Studies Of Trinuclear N₄ Schiff Base Complexes," International Journal of ChemTech Research, 5(1) 180-185, 2013.

[19] N. Turan and M. Sekerci, "Synthesis and Characterization of Co(II), Ni(II), Cu(II) and Cd(II) Complexes of Schiff Base Derived from 1,8-Diaminonaphthalene," Asian Journal of Chemistry, 21(9), 6939, 2009.

[20] R. K. Dubey, U. K. Dubey and C. M. Mishra, "Synthesis, physicochemical characterization of some Schiff base complexes Cr(III)," Indian Journal of Chemistry 47A, 1208-1212, 2008.

[21] N. Raman, J. D. Raja and A. Sakthivel, "Synthesis, spectral characterization of Schiff base transition metal complexes: DNA cleavage and antimicrobial activity studies," J. Chem. Sci., 119 (4), 303–310, 2007.

[22] S. K. Datta, E. R. T. Tiekink and M. Chaudhary, "Mono- and dinuclear oxovanadium (IV) compounds containing VO(ONS) basic core: synthesis, structure and spectroscopic properties," Polyhedron, 16(11), 1863-1871, 1997.