

Research Article

Synthesis and Studies on S-Triazine-Based Ligand and Its Metal Complexes

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Received 30 September 2012; Accepted 11 November 2012

Academic Editor: Rabindranath Mukherjee

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Tridentate chelate complexes of ML type (where M=Cu(II), Ni(II), and Co(II)) have been synthesized from triazine-based ligand 4,6-bis(5-mercapto-1,3,4-thiadiazol-amine)2-phenylamino-1,3,5-triazine (BMTDT). Microanalytical data, magnetic susceptibility measurements, IR, ¹H NMR, UV-vis, mass, and EPR spectral techniques were used to characterise the structure of chelates. The electronic absorption spectra and magnetic susceptibility measurements suggest that metal complexes show square pyramidal geometry. The electrochemical behavior of copper(II) complex is studied by cyclic voltammetry. All synthesized compounds may serve as potential photoactive materials as indicated from their characteristic fluorescence properties. The second harmonic generation efficiency (SHG) of the ligand and metal complexes has been found to be higher than that of urea and KDP. The antimicrobial activity of the ligand and metal(II) complexes against the species *Shigella*, *Chromobacterium*, *Staphylococcus aureus*, *Candida albicans*, and *Aspergillus niger* has been carried out and compared with the standard one.

1. Introduction

The rapid development of pathogen resistance to most of the known antibiotics is becoming a serious health problem [1]. One possible long-term solution is the development of agents that act on unexploited bacterial targets. S-triazine derivatives have received considerable attention due to their potent biological activity such as anticancer [2], estrogen receptor modulators [3], antivirals [4], and antimalarials [5, 6]. It has been reported that triazine derivatives possess potent antimicrobial activity [7, 8]. The compounds containing thiadiazole moiety have also been found to possess antitubercular and anti-inflammatory activities [9]. Hence, our aim is to synthesis a new derivative of triazine to enhance the pharmacological activities and nonlinear properties. Here, we present the synthesis, spectral characterization, bioscreening, nonlinear optical property, and fluorescence study of ligand derived from 2-amino-5-mercapto-1,3,4-thiadiazole-2-thiol and 2-phenylamino-1,3,5-triazine and its transition metal complexes.

2. Experimental

All chemicals were obtained from Aldrich Chemical & Co. and used without purification. They included carbon disulphide, hydrazine hydrate, cyanuric chloride, Cu(II) Cl₂·2H₂O, Co(II)Cl₂·6H₂O, and Ni(II)Cl₂·6H₂O. The solvents ethanol, DMF, and DMSO were used after purification by the standard method described in the literature. The UV-vis spectra of the ligand and its metal complexes were recorded using a JASCO V-530 Spectrophotometer. Fluorescence spectra were obtained on ELICO SL174 spectrofluorometer (Lady Doak College, Madurai) using DMSO as solvent. The IR spectra in KBr disc were recorded on a SHIMADZU FT-IR spectrophotometer at USIC, Madurai Kamaraj University, Madurai. Cyclic voltammetry measurements were carried out at room temperature in DMSO using a three-electrode cell containing a reference Ag/AgCl electrode, Pt wire auxiliary electrode, and glassy carbon working electrode with tetrabutylammoniumperchlorate (TBAP) as supporting electrolyte. The metal contents of the

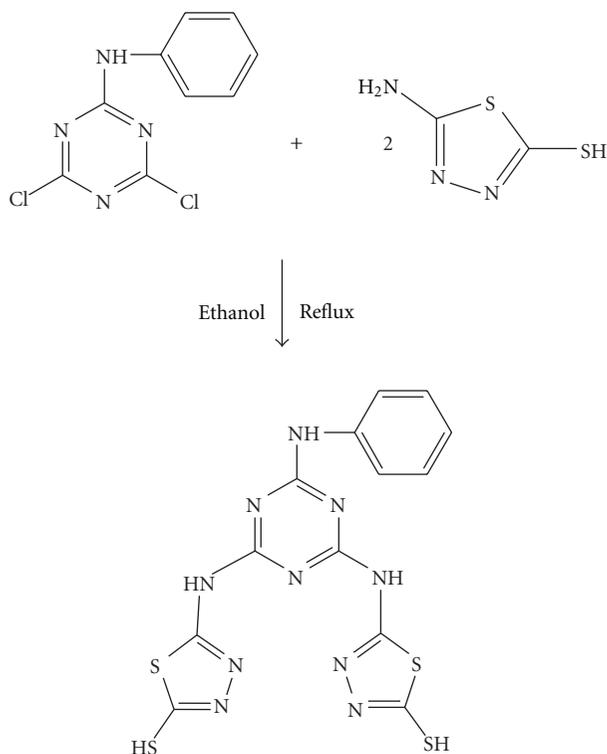


FIGURE 1: Scheme for the synthesis of ligand.

complexes were estimated by incinerating them to oxides [10]. ^1H NMR spectrum was recorded in CDCl_3 using a Bruker DRX-300, 300 MHz NMR spectrometer. EI mass was recorded by JEOL-GC MATE-2 at IIT, Madras, Chennai. EPR spectrum of copper(II) complex was recorded by VarianE-112 spectrometer at X-band, using TCNE with 100 KHz modulation frequency at SAIF, IIT, Bombay, India. Magnetic susceptibility of the complexes was measured by MSB mark 1 Sherwood UK at Thiagarajar College, Madurai. Effective magnetic moments were calculated using the formula $\mu_{\text{eff}} = (2.828\chi_M T)^{1/2}$, where χ_M is the corrected molar susceptibility. The diamagnetic corrections were made by Pascal's constant and $\text{Hg}[\text{Co}(\text{SCN})_4]$ was used as a calibrant. Molar conductance of the complexes (10^{-3}M) was measured in DMSO at room temperature using a Systronics conductivity bridge. All the complexes were screened for biological activity through the well-diffusion method. The SHG conversion efficiency of the ligand and its complexes were determined by the modified version of powder technique at IISc, Bangalore.

2.1. Synthesis of Ligand. 2.66 g (20 mmol) of 2-amino-5-mercapto-1,3,4-thiazole-2-thiol [11] in ethanol was refluxed with 2.40 g (10 mmol) of 2-phenylamino-4,6-dichloro-1,3,5-triazine [12] for 6 h. The white solid separated out was filtered, purified by recrystallisation from hot ethanol, and dried in vacuum over anhydrous calcium chloride.

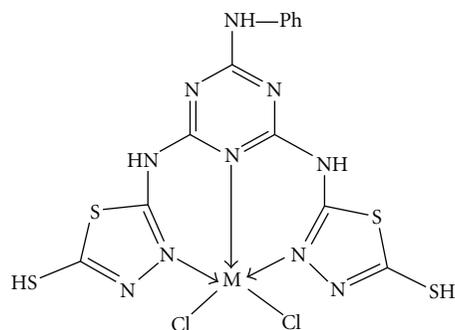


FIGURE 2: Proposed structure of the metal complexes $\text{M} = \text{Cu}(\text{II}), \text{Ni}(\text{II}), \text{and Co}(\text{II})$.

2.2. Synthesis of Metal(II) Complexes. An ethanolic solution of ligand (10 mmol) was refluxed with anhydrous metal salts MX_2 (10 mmol; $\text{M} = \text{Cu}(\text{II}), \text{Ni}(\text{II}), \text{Co}(\text{II})$) in 20 mL ethanol for 12 h. The characteristic colored complex precipitated was filtered, washed with ethanol and dried in vacuum.

2.3. Nonlinear Optical Properties of Ligand and Metal Complexes. The SHG efficiency of ligand and metal complexes was determined by the modified version of the powder technique developed by Kurtz and Perry [13]. The ligand was ground into powder and packed between two transparent glass slides. An Nd:YAG laser beam of wavelength 1064 nm was passed through the sample cell. The transmitted fundamental wave was absorbed by a copper(II) sulphate solution, which removes the incident 1064 nm light and Filter BG-38 removing any residual 1064 nm light. Interference filter band width is 4 nm and for central wavelength of 532 nm. Green light is finally detected by the photomultiplier tube and displayed on the oscilloscope. The second harmonic signal was detected by a photomultiplier tube and displayed on a storage oscilloscope. The efficiency of the sample was compared with microcrystalline powder of KDP and urea. The input energy used in this particular setup was 2.2 mJ/pulse.

3. Results and Discussion

All the complexes were obtained as solids by the reaction of ligand with anhydrous metal salts. The elemental analysis of the ligand and its metal complexes (Table 1) is in good agreement with the calculated ones. The ligand and its metal(II) complexes are stable at room temperature and soluble in common organic solvents such as DMSO, DMF, and acetone. Based on the elemental analysis and spectral studies, the coordination geometry has been assigned and is shown in Figures 1 and 2. The molar conductance of complexes in 10^{-3} molar DMSO solution is in the range $5.3\text{--}8.6 \text{ ohm}^{-1} \text{ cm}^2 \text{ mol}^{-1}$, which reveals the nonelectrolytic nature of the complexes [14].

TABLE 1: Physical, analytical, magnetic susceptibility, and molar conductance data of the ligand and its complexes.

Compound	m.p. (°C)	Calculated (found)%				λ_m ($\Omega^{-1} \text{ cm}^2 \text{ mol}^{-1}$)
		C	H	N	S	
BMTDT	110	35.87 (35.90)	2.28 (2.30)	32.23 (32.25)	29.43 (29.49)	—
Cu(BMTDT)Cl ₂	175	27.42 (27.46)	1.73 (1.76)	24.62 (24.64)	22.50 (22.53)	5.3
Ni(BMTDT)Cl ₂	184	27.72 (27.70)	1.73 (1.77)	24.83 (24.86)	22.75 (22.73)	8.6
Co(BMTDT)Cl ₂	210	27.81 (27.85)	1.73 (1.78)	25.04 (25.00)	22.57 (22.52)	6.7

TABLE 2: IR spectral data of the ligand and metal(II) complexes (cm^{-1}).

Compound	ν (C=N)	ν (C=N) triazine	ν (C-N) triazine	ν (M-Cl)	ν (M-N)
BMTDT	1646	1485	827	518	468
Cu(BMTDT)Cl ₂	1608	1456	810	553	464
Ni(BMTDT)Cl ₂	1602	1448	808	509	472
Co(BMTDT)Cl ₂	1604	1452	812	528	451

3.1. IR Spectra. The IR spectral data of the complexes (Table 2) usually provide a lot of valuable information on coordination mechanism. The ligand which exhibits a band at 1646 cm^{-1} is characteristic of (C=N) of thiadiazole ring. In complexes, this strong band is shifted to $1602\text{--}1608 \text{ cm}^{-1}$ region corresponding to (C=N) coordination to metal ion. The ligand shows a strong band at 827 cm^{-1} and 1485 cm^{-1} , which is characteristic of the ν (C-N) and ν (C=N) groups in S-triazine. This band shifting to lower frequency of $808\text{--}812 \text{ cm}^{-1}$ and $1448\text{--}1456 \text{ cm}^{-1}$ upon complexation indicates that triazine ring nitrogen is one of the coordinating atoms in the ligand [15]. In the far IR spectra of complexes, the weak bands appeared at $451\text{--}468 \text{ cm}^{-1}$ and $342\text{--}358 \text{ cm}^{-1}$ regions can be assigned to ν (M-N) and ν (M-Cl) vibrations [16], respectively, and confirm the interaction between metal and ligand.

3.2. Mass Spectra. The molecular ion peak for the ligand is observed at $434 m/z$ which is in good agreement with the suggested molecular formula $\text{C}_{13}\text{H}_{10}\text{N}_{10}\text{S}_4$. The fragmentation of the ligand is taken as a general scheme showing the main fragmentation paths involved and the fragmentation pattern is given in supplementary material available online at doi:10.1155/2012/301086.

3.3. Electronic Spectra. The electronic spectral data of metal(II) complexes are presented in Table 3. The electronic spectrum of Cu(II) complex showed three absorption bands at 12700 cm^{-1} , 15200 cm^{-1} , and 18500 cm^{-1} , which can be tentatively assigned to the transitions ${}^2\text{B}_1 \rightarrow {}^2\text{A}_1$ (ν_1), ${}^2\text{B}_1 \rightarrow {}^2\text{B}_2$ (ν_2), and ${}^2\text{B}_1 \rightarrow {}^2\text{E}$ (ν_3), respectively, are characteristic of square pyramidal geometry [17]. Such bands near 18656 cm^{-1} , 14815 cm^{-1} , and 10799 cm^{-1} have been observed for related copper(II) complexes with square pyramidal geometry [18, 19]. The copper(II) complex at 1.81 B.M. is interpreted as suggestive of a five-coordinate square pyramidal geometry [20]. The molar extinction coefficient for Cu(II) complex (ϵ) is in the range of several hundreds indicating the noncentrosymmetric nature of the complex. The electronic spectrum of Ni(II) complex

exhibits three d-d bands at 10000 cm^{-1} , 16474 cm^{-1} , and 23041 cm^{-1} and is assigned to the transitions ${}^3\text{B}_1(\text{F}) \rightarrow {}^3\text{E}(\text{F})$, ${}^3\text{B}_1(\text{F}) \rightarrow {}^3\text{A}_2(\text{P})$, and ${}^3\text{B}_1(\text{F}) \rightarrow {}^3\text{E}(\text{P})$, respectively, consistent with square pyramidal environment [21]. The spectra resemble those reported for five-coordinate square pyramidal nickel(II) complexes [22, 23]. The Co(II) complex displayed three absorption bands at 9803 cm^{-1} , 16420 cm^{-1} , and 20000 cm^{-1} which can be assigned to the transitions ${}^4\text{A}_2 + {}^4\text{E} \rightarrow {}^4\text{B}_1$ (ν_1), ${}^4\text{A}_2 + {}^4\text{E} \rightarrow {}^4\text{E}(\text{P})$ (ν_2), and ${}^4\text{A}_2 + {}^4\text{E} \rightarrow {}^4\text{A}_2(\text{P})$ (ν_3), respectively, which are characteristic of square pyramidal geometry [24]. Cobalt(II) and Nickel(II) complexes have magnetic moment values 4.69 and 3.02 B.M., respectively, also further confirms the square pyramidal geometry. The molar absorptivity (ϵ) of the above complexes is in the order of 10^2 M cm^{-1} indicates the geometry of the molecule.

3.4. Electrochemical Behavior. Cyclic voltammogram of copper(II) complex in DMSO recorded at room temperature using TBAP as the supporting electrolyte from the potential range 1 to -0.6 V range displays a pair of oxidation-reduction peaks at 0.027 V and -0.400 V (Figure 8). The redox peak potential separation (ΔE_p) of 427 mV suggests that the Cu(II)/Cu(I) redox couple was irreversible in nature [25]. The ratio of anodic-to-cathodic peak current for the complex ($I_{pc}/I_{pa} \approx 1$) corresponding to a simple one-electron process. The reason for the irreversibility observed for the reductive response of the complex may be due to a short-lived reduced state of the metal ion or due to the oxidative degradation of the ligand [26].

3.5. EPR Spectra. A powder EPR spectrum of Copper(II) complex was recorded at 300 K and 77 K and is appeared slightly broadened (Figure 7) with $g_{\parallel} = 2.156$ and $g_{\perp} = 2.066$ —characteristic of square pyramidal geometry [27]. The observed g_{\parallel} value for the complex is less than 2.3, suggesting significant covalent character of the metal-ligand bond in agreement with the observation of Kivelson and Neiman. By observing g -values, it is clear that $g_{\parallel} > g_{\perp} > g_e$, suggesting that $d_{x^2-y^2}$ orbital is the ground state and

TABLE 3: Electronic spectral data of ligand and metal(II) complexes.

Compound	Absorption (cm ⁻¹)	Band assignment	Geometry	μ_{eff} (BM)
Cu(BMTDT)Cl ₂	12706	² B ₁ → ² A ₁	Square pyramidal	1.81
	15220	² B ₁ → ² B ₂		
	18518	² B ₁ → ² E		
Ni(BMTDT)Cl ₂	10000	³ B ₁ (F) → ³ E(F)	Square pyramidal	3.02
	16474	³ B ₁ (F) → ³ A ₂ (P)		
	23041	³ B ₁ (F) → ³ E(P)		
Co(BMTDT)Cl ₂	9803	⁴ A ₂ + ⁴ E → ⁴ B ₁	Square pyramidal	4.69
	16420	⁴ A ₂ + ⁴ E → ⁴ E(P)		
	20000	⁴ A ₂ + ⁴ E → ⁴ A ₂ (P)		

TABLE 4: Spin Hamiltonian parameter for Cu(II) complex.

Complex	g_{\parallel}	g_{\perp}	A_{iso}	g_{iso}	A_{\parallel} (10 ⁻⁴ cm)	A_{\perp} (10 ⁻⁴ cm)	α^2	β^2	$g_{\parallel}/A_{\parallel}$
Cu(BMTDT)Cl ₂	2.156	2.066	95.2	2.095	187.25	49.2	0.741	0.62	115.3

the d⁹ configuration is (e_g)⁴(a_{1g})²(b_{2g})²(b_{1g})¹. This is also supported from the value of the exchange interaction term G , estimated from the expression,

$$G = \frac{g_{\parallel} - 2.0023}{g_{\perp} - 2.0023} \quad (1)$$

If $G > 4.0$, the local axes are aligned parallel or only slightly misaligned. If $G < 4.0$, significant exchange coupling is present and the misalignment is appreciable. The observed value for the exchange interaction parameter for the copper complex ($G < 4.0$) suggests considerable interaction in the solid state. The spin-orbit coupling constant, λ calculated using the relations, $g_{\text{ave}} = 1/3[g_{\parallel} + g_{\perp}]$, and $g_{\text{ave}} = 2(1 - 2\lambda/10Dq)$, is less free Cu(II) (-828), which also supports covalent character of metal-ligand bond in the complex. The covalency parameter α^2 is calculated using the following:

$$\alpha^2 = \frac{A_{\parallel}}{P} + (g_{\parallel} - 2.0023) + \frac{3}{7(g_{\perp} - 2.0023)} + 0.04, \quad (2)$$

$\alpha^2 = 0.5$ indicates complete covalent bonding, while the value of $\alpha^2 = 1$ suggests complete ionic bonding (Table 4). The observed values of α^2 of the complex are less than unity, which indicates that the complex has some covalent character [28].

3.6. Fluorescence Studies. The emission spectra of the ligand and its metal(II) complexes were studied (Table 5), the ligand exhibits a broad and strong greenish-blue emission bands with wavelength 522 nm. The Cu(II), Ni(II), and Co(II) complexes exhibit emission bands at 681 nm, 580 nm, and 679 nm, respectively (Figures 3, 4, 5, and 6). Significant differences in the positions of emission maximum of ligand and its complexes establish the coordination of the metal ion to the ligand. Enhancement of fluorescence through complexation is much interesting as it opens up the opportunity for photochemical applications of these complexes. The

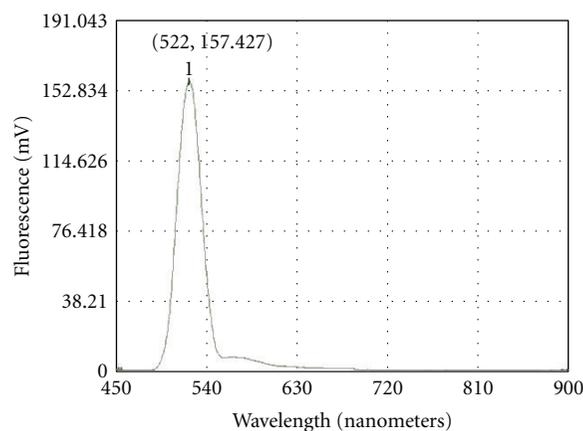


FIGURE 3: Fluorescence spectrum of ligand.

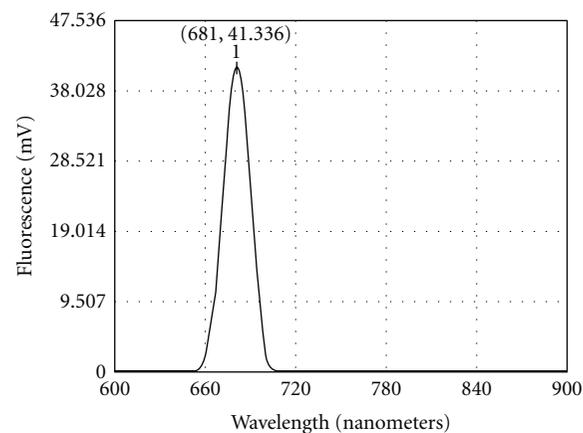


FIGURE 4: Fluorescence spectrum of Cu(II) complex.

emission is readily observed for complexes with the naked eyes under UV light, whereas the ligand exhibits quite weak fluorescence.

TABLE 5: Fluorescence parameters of ligand and its metal(II) complexes.

Compound	Excitation wavelength λ_{abs} (nm)	Fluorescence wavelength λ_{emi} (nm)
BMTDT	455	522
Cu(BMTDT)Cl ₂	657	681
Ni(BMTDT)Cl ₂	496	580
Co(BMTDT)Cl ₂	609	679

TABLE 6: Biological activity of ligand and metal(II) complexes (zone of inhibition in mm^a).

Compound	<i>Shigella</i>	<i>Chromobacterium</i>	<i>S. aureus</i>	<i>C. albicans</i>	<i>A. niger</i>
BMTDT	12	12	6	8	R
Cu(BMTDT)Cl ₂	15	15	10	11	16
Ni(BMTDT)Cl ₂	17	12	15	12	19
Co(BMTDT)Cl ₂	14	14	12	14	20
Amikacin	20	18	17	—	—
Ketoconazole	—	—	—	25	23

^a Each value is observed within the error limits of ± 1 .

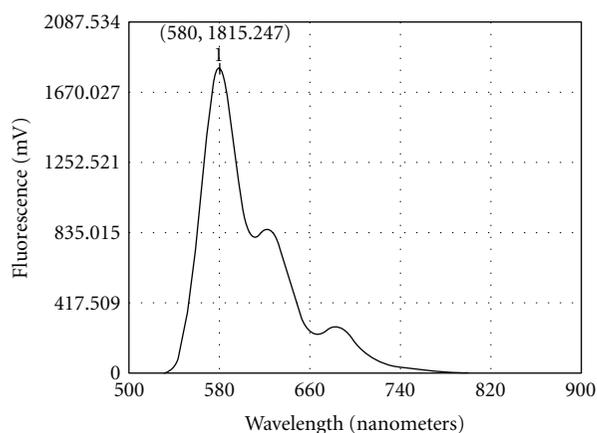


FIGURE 5: Fluorescence spectrum of Ni(II) complex.

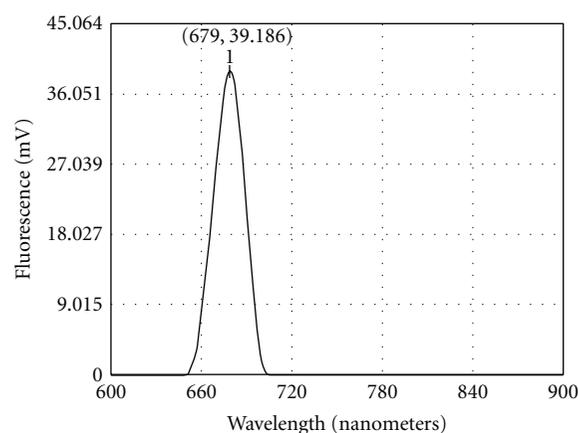


FIGURE 6: Fluorescence spectrum of Co(II) complex.

3.7. NLO Property. The second harmonic generation (SHG) efficiency of the ligand shows much higher efficiency than KDP and urea. This may be due to the presence of thiadiazole and triazine nucleus. In general, molecule with delocalized π -electron system can have large nonlinear polarizabilities [29] and it has been generally understood that the molecular non linearity can be enhanced by systems with strong donor and acceptor groups [30]. The second harmonic generation efficiency clearly reveals that the ligand possess 0.4 times better activity than that of urea and 2.4 times more active than KDP. Continuing our research on NLO activity to Cu(II), Ni(II), and Co(II) complexes, the result shows that the complexes are 0.7, 0.6, and 0.5 times better activity than urea, respectively, and 3.5, 3.2, and 2.9 times higher activity than KDP, respectively.

3.8. Biological Activity. The ligand and its metal(II) complexes were evaluated for antibacterial activity and antifungal activity by using well-diffusion method (Table 6). Amikacin and Ketoconazole were taken as the standard antibiotics and

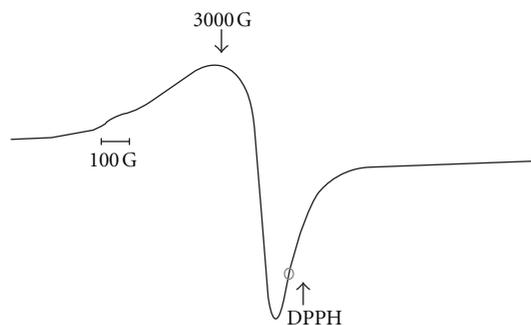


FIGURE 7: EPR spectrum of Cu(II) complex.

antifungi. Antimicrobial activity studies were performed in triplicate and the average was taken as the final reading. Error limits are indicated in the respective table. *In vitro* antifungal activity data of the ligand and its metal(II) complexes against tested organisms displayed significant activity with wide range of variation. It is found that all the derived compounds

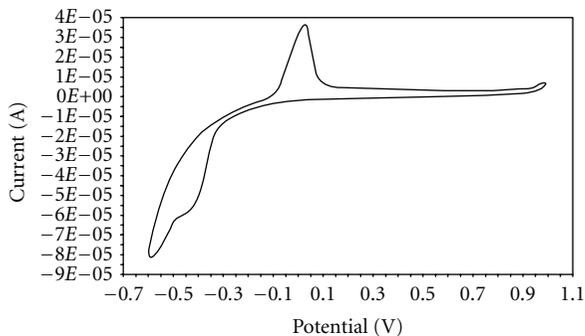


FIGURE 8: Cyclic voltammogram of Cu(II) complex.

exhibited higher activity against all the organisms but still lower than the standard. Ligand is inactive towards *A. niger* but the metal complexes show higher activity against the same organism compared to standard.

In vitro antibacterial activity data reveals that all the newly synthesized compounds displayed moderate to significant activity in comparison to standards. The ligand and Ni(II) complexes are equipotent against *Chromobacterium* and the rest of the compounds exhibited higher activity against the same strain. It is found that Ni(II) complex displayed significant activity against *Shigella* and remaining compounds are substantially active. The ligand exhibited moderate activity against *S. aureus* and all the metal complexes displayed significant activity compared to ligand. Such an increased activity of the metal(II) complexes over the ligand can be explained on the basis of Overton's concept and the Chelation theory [31, 32]. On chelation, the polarity of the metal ion will be reduced to a greater extent due to the overlap of the ligand orbital and partial sharing of positive charge of the ligand orbital and donor groups. The mode of the action of compounds may involve the formation of hydrogen bond with the active center of cell constituents resulting in interference with normal cell process.

4. Conclusion

The synthesized tridentate ligand undergoes coordination through triazine ring nitrogen and thiadiazole ring nitrogen with metal(II) ions. The analytical and spectral studies confirm the bonding of ligand to metal(II) ions and it confirms all the metal(II) complexes possess square pyramidal geometry. Electrochemical study of the copper(II) complex implies the redox reaction is one electron transfer with irreversible character. The ligand and its metal complexes were found to exhibit appreciable nonlinear property in comparison with KDP and urea. Hence, the compounds can be utilized for further NLO investigations. The ligand and its metal complexes are fluorescent in nature. The ligand and its metal(II) complexes were potentially active towards all microbial strains. The fungal studies of all the metal complexes against *A. niger* show promising results. Such increased activity of the metal complex is of tremendous importance in the pharmaceutical field.

Acknowledgments

The authors thank the Management of Thiagarajar College, Madurai and one of the authors (P. Tharmaraj) thanks the Defence Research and Development Organization (DRDO) for providing financial support and SAIF, IIT Bombay, IISc, Bangalore for providing analytical facilities.

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