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Microwave synthesis, spectral, thermal and antimicrobial activities of Co(II), Ni(II) and Cu(II) metal complexes with Schiff base ligand

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ABSTRACT

The complexes of tailor made ligands with life essential metal ions may be an emerging area to answer the problems of multi drug resistance (MDR). Some novel Schiff base metal complexes of Co(II), Ni(II) and Cu(II) derived from 4-chlorobenzylidene-2,6-dichloro-4-nitroaniline (CDN) have been synthesized by conventional as well as microwave methods. These compounds have been characterized by elemental analysis, FT-IR, FAB-mass, molar conductance, electronic spectra, ESR, magnetic susceptibility, and thermal analysis. The complexes are coloured and stable in air. Analytical data revealed that all the complexes exhibited 1:2 (metal: ligand) ratio with coordination number 4. FAB-mass and thermal data show degradation pattern of the complexes. The crystal system, lattice parameter, unit cell volume and number of molecules in unit cell in the lattice of complexes have been determined by XRD analysis. XRD patterns indicate crystalline nature for the complexes. The Schiff base and metal complexes show a good activity against the bacteria; *E. coli, S. aureus, S. fecalis* and fungi *A.niger, T. polysporum, C. albicans.* The antimicrobial results also indicate that the metal complexes are better antimicrobial agents as compared to the Schiff base.

Keywords: *microwave synthesis; N donor; thermal analyses; XRD; biological activity*

1. INTRODUCTION

Most of the major classes of pharmaceutical agents contain examples of metal compounds which are in current clinical use and new areas of application are rapidly emerging. It is well known that N atom play a key role in the coordination of metals at the active sites of numerous metallobiomolecules. The application of Schiff bases and their metal complexes in the field of industry, agriculture and medicine are well known. The biological potential of the aldehydes *viz.* 4-chlorobenzaldehyde after condensation with amines 2,6-dichloro-4-nitroaniline might get enhanced, so such Schiff base has been selected for complexation with the metal ions *viz.* Co(II), Ni(II) and Cu(II). The coordination chemistry with its growing list of applications, has paved the way to the preparation of new compounds which may possess some definite predetermined properties and thus show promising prospects in the realm of medicine and molecular biology *viz.* designing of metal complexing drugs (inorganic drugs) and metallotherapy. They used as models for biological systems and find applications in biomimetic catalytic reactions. The most widely studied metal in this respect

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is copper(II) which has proved to be beneficial in diseases such as tuberculosis, gastric ulcers, rheumatoid arthritis and cancers [1-5]. Microwave-assisted synthesis is a branch of green chemistry. Microwave irradiated reactions are offering reduced pollution, low cost and offer high yield together with simplicity in processing and handling. The basis of this technique of synthesis is much faster with high yields compared to conventional methods. In conventional methods, the yield is lower as compared to microwave irradiation, demonstrating the effect of microwave irradiation which is not purely thermal. Microwave irradiation facilitates the polarization of the molecule under irradiation causing rapid reaction to occur. The salient features of microwave approach are shorter reaction times, simple reaction conditions and enhancements in yields [6-10]. Reports on the synthesis of metal complexes by microwave methods have been comparatively less. In the present paper, we have described the coordination behavior of Schiff base derived from the condensation of 4-chlorobenzaldehyde with 2,6-dichloro-4-nitroaniline towards some transition elements, which may help in more understanding of the mode of coordination of ligand towards metals. For this purpose the complexes of Co(II), Ni(II) and Cu(II) ions with CDN have been synthesized by both conventional as well as microwave methods and characterized by the various physicochemical and spectral analyses. The metal complexes formed with new ligand may be used as precursors for the synthesis of new compounds. Some of which them may exhibit interesting physical and chemical properties and potentially useful biological activities.

2. EXPERIMENTAL SECTION

2.1. Materials and instrumentation. All the used chemicals and solvents were of A. R. grade from Sigma Aldrich. Elemental analyses were performed on an Elemental Vario EL III Carlo Erba 1108 analyzer. FAB-mass spectra were recorded on a JEOL SX 102/DA 6000 Mass Spectrometer using argon/xenon (6 kV, 10 mA) as the FAB gas. The accelerating voltage was 10 kV and the spectra were recorded at room temperature. Electronic spectra (in DMSO) were recorded on Perkin Elmer Lambda-2B-spectrophotometer. Molar conductance measurements were conducted using 10^{-3} M solutions of the complexes in DMSO on Elico-CM 82 Conductivity Bridge at room temperature. Magnetic susceptibility measurements were carried out on a Gouy balance at room temperature using $Hg[Co(SCN)_4]$ as the calibrant. Diamagnetic corrections were applied in compliance with Pascal's constant. FT-IR spectra were recorded in KBr medium on a Perkin Elmer RX1 spectrophotometer in wave number region 4000-400 cm^{-1}. X-band EPR spectra were recorded on a Varian E-112 spectrometer at room temperature operating at the X-band region with 100 kHz modulation frequency, 5 mw microwave power and 1 G modulation amplitude using TCNE as the internal standard. Thermogravimetric analysis was carried out under atmospheric condition with a heating rate $10^{o}C$ min^{-1} on TGA Q500 universal V4.5A TA instrument. Powder X-ray diffraction (XRD) patterns were recorded on a RINT2000 wide angle goniometer. X-ray diffractometer, operated at 40 kV and 30 mA generator using the CuKα line at 1.5405Å as the radiation sources. Sample was scanned between 5° to 70° (2θ) at 25 °C. X-ray crystal system has been worked out by trial and error methods for finding the best fit between observed and calculated $\sin^2\theta$. The unit cell volume (V), number of molecules in unit cell in the lattice (Z), calculated and observed density (D) and particle size have been calculated. Microwave assisted synthesis were carried out in open glass vessel on a modified microwave oven model 2001 ETB with rotating tray and a power source 230 V, microwave energy output 800W and microwave frequency 2450 MHz. A thermocouple device was used to monitor the temperature inside the vessel of the microwave. The microwave reactions were performed using on/off cycling to control the temperature.

2.2. Antimicrobial Activity. The *in-vitro* biological activity of the investigated Schiff base and its metal complexes was tested against the bacteria *Escherichia coli, Staphylococcus aureus* and *Streptococcus fecalis* by disc diffusion method using nutrient agar as medium and Gentamycin as control. The antifungal activities of the compounds were also tested by the Well diffusion method against the fungi *Aspergillus niger, Trichoderma polysporum* and *Candida albicans*, on potato dextrose agar as the medium and nystatin as control. Each of the compounds was dissolved in DMSO and solutions of the concentrations (25, 50 and 100 ppm) were prepared separately. In a typical procedure, a well was made on agar medium inoculated with microorganism. The well was filled with the test solution using a micropipette and the plate was incubated 24 h for bacteria at 37°C and 72 h for fungi at 30°C. During this period, the test solution diffused and the growth of the inoculated microorganism was affected. The inhibition zone was developed, at which the concentration was noted.

2.3. Conventional synthesis of the Ligand. The ligand (CDN) (Figure 1) has been synthesized by the addition of the methanolic solution of 4-chlorobenzaldehyde (0.01 mol) to the methanolic solution of 2,6-dichloro-4-nitroaniline (0.01 mol) in 1:1 ratio. The reaction mixture is refluxed on a water bath for about 4 hour. The brownish coloured product is recrystallized with ethanol and petroleum ether and purity is checked by TLC using silica gel G (yield: 74%).

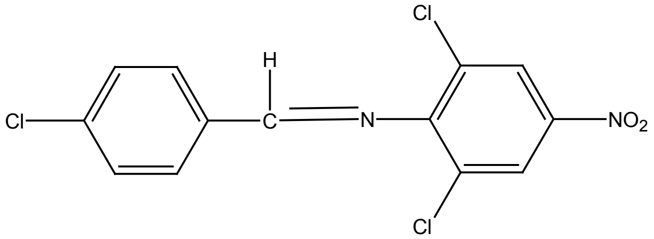


Figure 1: Structure of CDN (Schiff base).

2.4. Microwave method for the Synthesis of Schiff base. The equimolar (1:1) ratio of 4-chlorobenzaldehyde and 2,6-dichloro-4-nitroaniline was mixed thoroughly in a grinder. The reaction mixture was then irradiated by the microwave oven using 3-4 ml solvent. The reaction was completed in a short time (4.2 min). The resulting product was then recrystallized with ethanol, finally dried under reduced pressure over anhydrous $CaCl_2$ in a desiccator. The progress of the reaction and purity of the product was monitored by TLC using silica gel G (yield: 89%).

2.5. Conventional synthesis of the metal complexes. Metal complexes have been synthesized by the addition of the methanolic solution of the appropriate metal salts $CoCl_2.6H_2O$, $NiCl_2.6H_2O$ and $CuCl_2.2H_2O$ to a methanolic solution of Schiff base 4-chlorobenzylidene-2,6-dichloro-4-nitroaniline in 1:2 ratio and the mixture is refluxed on water bath for about 6-8 hours. The refluxate is kept overnight. The resulting coloured precipitated complex is washed with ethanol followed by petroleum ether and dried under reduced pressure over anhydrous $CaCl_2$ in a dessicator (yield: 65-68%).

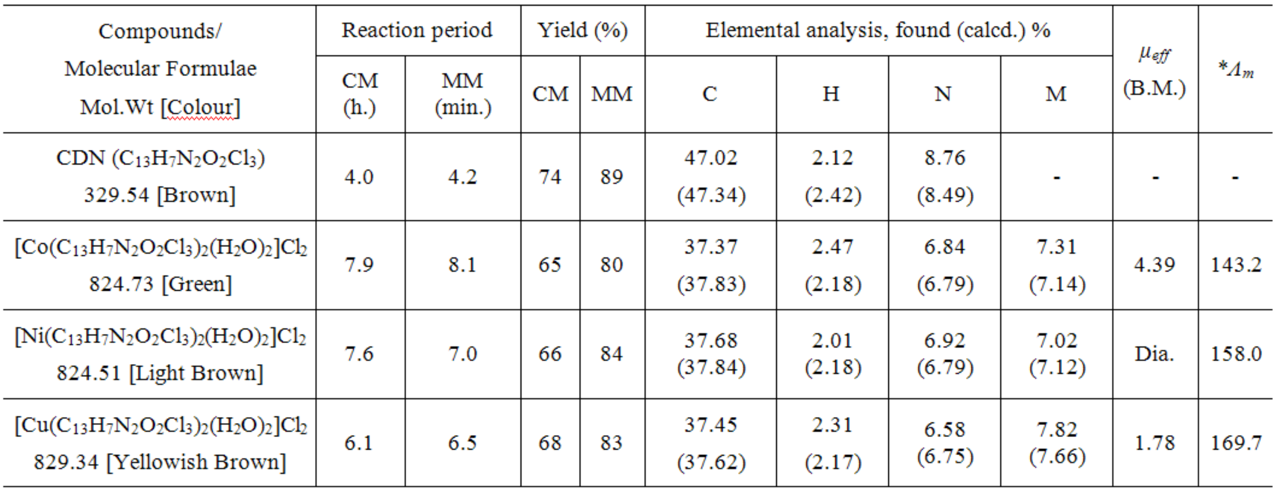
2.6. Microwave method for the Synthesis of metal complexes. The ligand and the metal salt were mixed in 1:2 (metal:ligand) ratio in a grinder. The reaction mixture was then irradiated in the microwave oven using 3-4 ml solvent. The reaction was completed in a short time (6-8 min). The resulting product was then recrystallized with ethanol and ether and finally dried under reduced pressure over anhydrous $CaCl_2$ in a desiccator. The progress of the reaction and purity of the product was monitored by TLC using silica gel G (yield: 80-84%).

3. RESULTS SECTION

As a result of microwave assisted synthesis, it was observed that the reaction was completed in a short time with higher yields compared to the conventional method. In the microwave method homogeneity of reaction mixture was increased by the rotating of reaction platform tray. The confirming of the results was also checked by the repeating of the synthesis process.

All the metal complexes are coloured, solid and stable towards air and moisture at room temperature. They decompose on heating at high temperature and more or less soluble in common organic solvents. Comparative study results obtained by microwave assisted synthesis; versus conventional heating method is that some reactions which required 4-8 h. by conventional method, was completed within 4-8 min. by the microwave irradiation technique and yields have been improved from 55-74% to 80-89%. The comparison study data of microwave and conventional methods, with analytical and physical data of the compounds are given in the Table 1. Analytical date show that metal complex have 1:2 (metal:ligand) stoichiometry. The molar conductance in DMSO suggests the uni-bivalent electrolytic nature of all the three complexes.

Table 1: The comparative results of conventional and microwave methods, analytical, physical data and magnetic moment values of the compounds



Compounds/ Molecular Formulae Mol.Wt [Colour]	Reaction period		Yield (%)		Elemental analysis, found (calcd.) %				μ_{eff} (B.M.)	$*\Lambda_m$
	CM (h.)	MM (min.)	CM	MM	C	H	N	M		
CDN ($C_{13}H_7N_2O_2Cl_3$) 329.54 [Brown]	4.0	4.2	74	89	47.02 (47.34)	2.12 (2.42)	8.76 (8.49)	-	-	-
$[Co(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ 824.73 [Green]	7.9	8.1	65	80	37.37 (37.83)	2.47 (2.18)	6.84 (6.79)	7.31 (7.14)	4.39	143.2
$[Ni(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ 824.51 [Light Brown]	7.6	7.0	66	84	37.68 (37.84)	2.01 (2.18)	6.92 (6.79)	7.02 (7.12)	Dia.	158.0
$[Cu(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ 829.34 [Yellowish Brown]	6.1	6.5	68	83	37.45 (37.62)	2.31 (2.17)	6.58 (6.75)	7.82 (7.66)	1.78	169.7

CM = Conventional method, time in hours; MM = Microwave method, time in minutes

$*\Lambda_{m} = (\Omega^{-1}\ cm^2 mol^{-1})$

3.1. FAB-mass spectra. The FAB-mass spectrum of $[Cu(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ complex has been recorded. The spectrum of the complex $[Cu(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ exhibited the molecular ion (M^+) peak at *m/z* 830 suggesting the monomeric nature of the complex. Other important peak observed at *m/z* 758, 720, 510, 415, 265, 196 and 95 correspond to fragment species of different groups. The *m/z* value 830 correspond to composition of $[Cu(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ and 95 correspond to copper metal with some coordinated-N ligand moiety [11].

3.2. IR spectra. In order to get data conceiving the ligand way of coordination to the metal ions, the IR spectra on the 400-4000 cm^{-1} range have been carried out. Characteristic peaks in the spectra of the ligand and complexes were considered and compared. A sharp band at 1622 cm^{-1} in the IR spectrum of the CDN ligand is owing to >C=N group. This band shifts down by 10-15 cm^{-1} in complexes, indicating coordination through azomethine nitrogen. In Schiff base, the nitro and two chloro groups at 4 and 2, 6 position/para and ortho position, are present in one part (2,6-dichloro-4-nitroaniline) of the ligand. The nitro group and chloro groups are electron withdrawing and have negative inductive effect (-I). This state leaves impact on the basicity of the ligand (Schiff base). Here, the Schiff base ligand has been considered to behave as monodentate ligand. The IR spectra of complexes exhibited new bands around 3359±8 and 740±5 (rocking), assignable to coordinated water molecules. The new band at 480±5 cm^{-1} in the complexes have tentatively been assigned to υ(M-N) [12-14].

3.3. Electronic spectra and magnetic moment. The nature of the ligand field around the metal ion has been deduced from the electronic spectra. The electronic spectrum of Co(II) complex of CDN shows one band at 16000 cm^{-1} (υ_3) which has been assigned to $^4A_2 \rightarrow ^4T_1(P)$ transition. Its magnetic moment is 4.39 B.M. On the basis of parameters, the tetrahedral geometry has been suggested for this complex. Nickel(II) complex of CDN exhibits three bands at 13831, 18580, 22458 cm^{-1}; these are tentatively assigned to $^1A_{1g} \rightarrow ^1E_g$ (υ_1) $^1A_{1g} \rightarrow ^1B_{2g}$ (υ_2) and $^1A_{1g} \rightarrow ^1B_{1g}$ (υ_3) transitions respectively. It is a diamagnetic complex, therefore square planar geometry has been suggested. Copper(II) complex of CDN gives bands at 14206 and 17992 cm^{-1}. These may tentatively be assigned to $^2B_{1g} \rightarrow ^2B_{2g}$ and $^2B_{2g} \rightarrow ^2E_g$ transitions. Its magnetic moment is 1.78 BM. This favours the square planar geometry [15-16].

3.4. ESR spectra. The X-band ESR spectrum of Cu(II) complex (Figure 2) was recorded in the solid state at room temperature and their $g_{\parallel}$, $g_{\perp}$, Δg, g_{av} and G have been calculated. The values of ESR parameters $g_{\parallel}$, $g_{\perp}$, g_{av}, Δg and G for Cu(II) complex of CDN are 2.2452, 2.1921, 2.2098, 0.0531 and 1.2793 respectively.

The trend $g_{\parallel} > g_{\perp} > g_e$ (2.0023) shows that the unpaired electron is localized in $d_{X^2-Y^2}$ orbital in the ground state of Cu(II) and spectra are characteristic of axial symmetry. The exchange coupling interaction between two Cu(II) ions is explained by Hathaway expression $G = (g_{\parallel}-2.0023)/(g_{\perp}-2.0023)$. According to Hathaway, if the value G is greater than four (G>4.0), the exchange interaction is negligible; whereas when the value of G is less than four (G<4.0) a considerable exchange coupling is present in solid complex. The G values for the Cu(II) complex are less than four indicating, considerable exchange interaction in the complexes [17, 18].

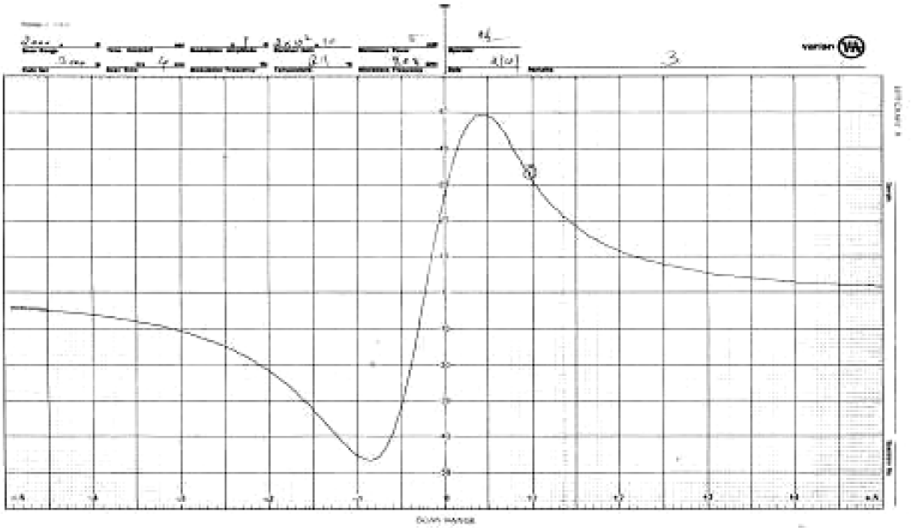


Figure 2: ESR spectrum of $[Cu(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ complex.

3.5. Thermal analyses. The thermal behavior of metal complex shows the degradation pattern of the complex. Thermal studies of complexes have been recorded in oxygen atmosphere. Thermal analysis data of complexes are given in Table 2.

A careful analysis of thermogram of $[Co(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ (Figure 3) indicates that complex compound is stable upto 160°C. Above this temperature a weight loss has been observed corresponding to two coordinated water molecules (Remaining wt%, Obs/Calc., 94.5/95.6). After 220 °C, a weight loss at faster rate occurs upto 340 °C, indicating decomposition of non-coordinated part of the ligand (Remaining wt%, Obs./Calc., 21.2/20.2). The decomposition of remaining ligand moiety occurs between 340-540 °C, above 540 °C a horizontal curve has been observed suggesting the ultimate pyrolysis product as metal oxide (Remaining wt%, Obs./Calc., 10.5/9.9). The thermogram of $[Ni(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ complex (Figure 4) does not loss weight upto temperature 140°C, indicating the absence of lattice water molecules. An elimination of two coordinated molecules of water has been observed between the temperature range 140°C-260°C (Remaining wt%, Obs./Calc., 95.1/95.6). Above this temperature (260°C), a gradual mass loss has been observed in general upto 300°C, indicating decomposition of non-coordinated part of the ligand (Schiff base) (Remaining wt%, Obs./Calc., 22.7/20.1). Above 300°C an inflection occurs in the curve and loss in weight progress upto 490°C, beyond which there lies a zone of constant weight corresponding to metal oxide as an ultimate pyrolysis product (Remaining wt%, Obs./Calc., 11.2/9.1) [16, 20].

Table 2: Thermal analyses data of complexes

Complexes	Temperature Range (°C)	Mass Loss (%)		Corresponding mass loss
		Obs.	Calc.	
Co(II)-CDN Complex	160-220	5.5	4.4	Two coordinated water molecules
	220-340	78.8	79.8	Non-coordinated part of the ligand
	340-540	89.5	90.1	Coordinated part of the ligand
Ni(II)-CDN Complex	140-260	4.9	4.4	Two coordinated water molecules
	260-300	77.3	79.9	Non-coordinated part of the ligand
	300-490	88.8	90.9	Coordinated part of the ligand

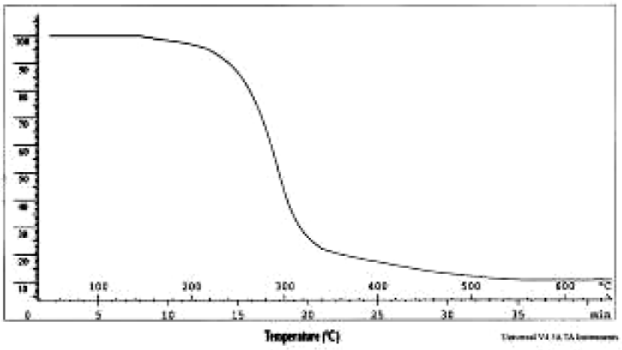


Figure 3: Thermogram of $[Co(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ complex.

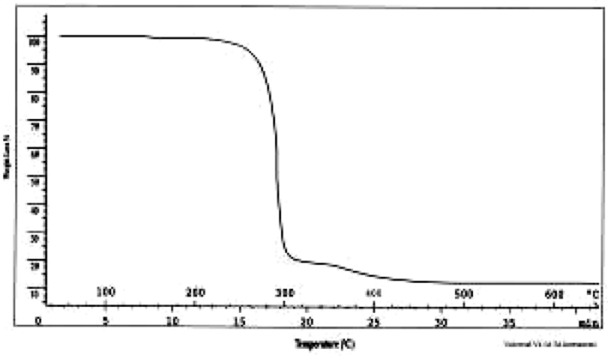


Figure 4: Thermogram of $[Ni(C_{13}H_7N_2O_2Cl_3)_2(H_2O)_2]Cl_2$ complex.

3.6. Powder X-ray diffraction. X-ray diffraction was performed of metal complexes. The XRD patterns indicate crystalline nature for the complexes. X-ray powder diffractogram of the complexes were recorded using CuKα as source in the range 5^{o}-70^{o} (2θ), X-ray crystal system has been worked out by trial and error methods for finding the best fit between observed and calculated $\sin^2\theta$. The X-ray powder diffractogram of nickel(II) complex of CDN crystallized in cubic system. Complex has recorded 12 reflections with maxima at d = 4.2025 Å. $\text{Sin}^2\theta$ and hkl values for different lattice planes have been calculated. Crystal data for complex, a = b = c = 20.02 Å, V = 8024.02 Å^3, Z = 9, $D_{(obs)}$ = 1.5344 g/cm^3, $D_{(cal)}$ = 1.6452 g/cm^3 and particle size = 13.24 nm. Observed and calculated values of density and $\sin^2\theta$ show good agreement. The X-ray powder diffractogram of Cu(II) complex of CDN was also recorded. Data reveals that the complex has crystallized in cubic system. Crystal data for Cu(II) complex are as: a = b = c = 37.99 Å, V = 5482.69 Å^3, Z = 8, $D_{(obs)}$ = 1.765 g /cm^3, $D_{(cal)}$ = 1.709 g /cm^3 and particle size = 11.65 nm [21].

3.7. Antimicrobial activity. The *in-vitro* Antimicrobial activity of the synthesized Schiff base ligands and their corresponding metal complexes on selected bacteria *Escherichia coli, Staphylococcus aureus* and *Streptococcus fecalis* and two fungi *Aspergillus niger, Trichoderma polysporum* and *Candida albicans* was carried out. All of the tested compounds showed good biological activity against microorganism. On comparing the biological activity of the Schiff base and its metal complexes with the standard bactericide and fungicide, it is show that the some metal complexes have good activity as compared to the standard but all the complexes are more active than their respective ligands and thus indicated that the complexation to metal enhances the activity of the ligand. Increase the activity due to some factors *viz.* solubility, lipophilicity/ hydrophilicity, conductivity and M-L bond length. [22-24].

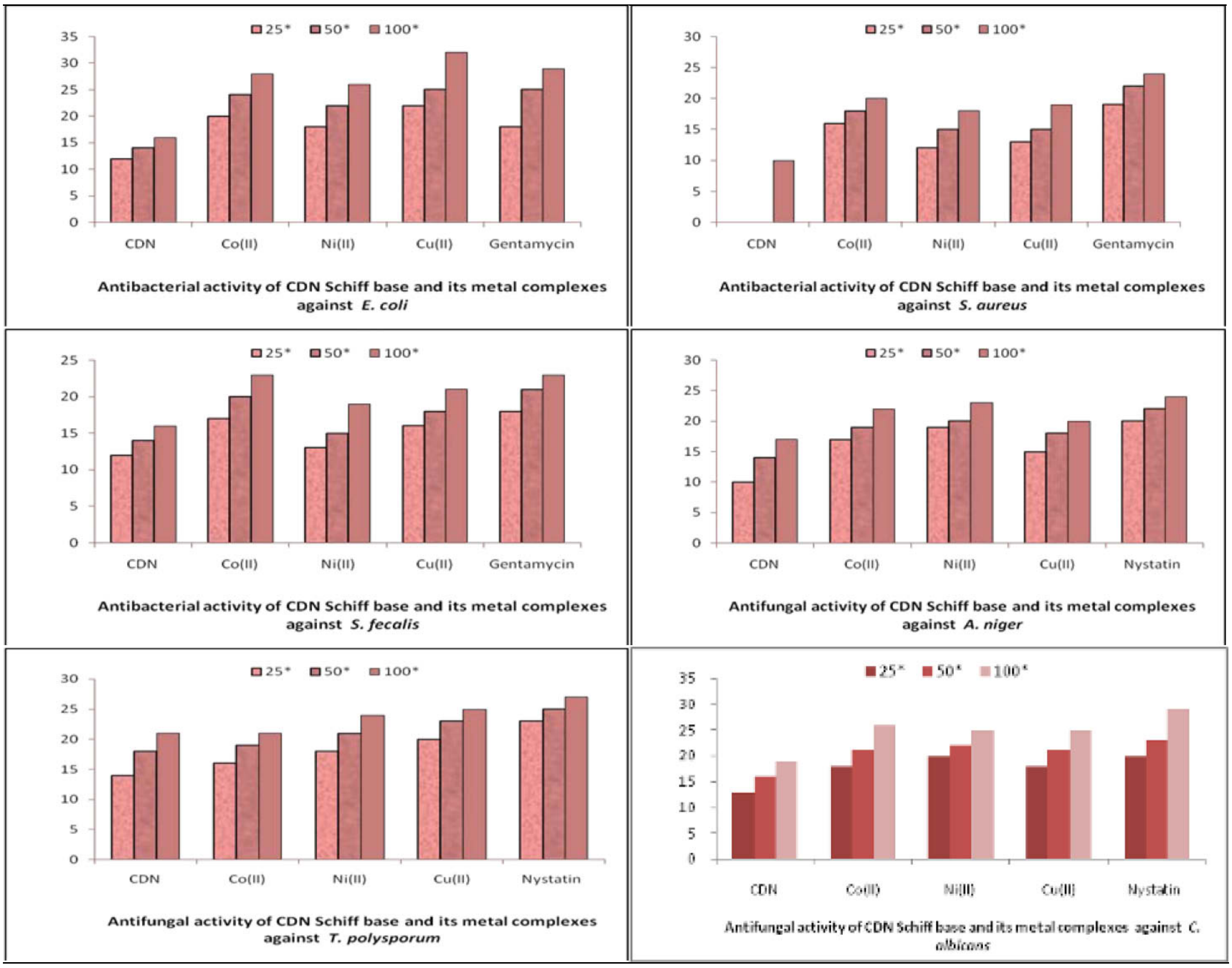
Table 3: Antimicrobial data of the investigated compounds

Compound	Antibacterial									Antifungal								
	E. coli			*S. aureus*			*S. fecalis*			*A. niger*			*T. polysporum*			*C. albicans*		
	25*	50*	100*	25*	50*	100*	25*	50*	100*	25*	50*	100*	25*	50*	100*	25*	50*	100*
CDN	12	14	16	-	-	10	12	14	16	10	14	17	14	18	21	13	16	19
Co(II)	20	24	28	16	18	20	17	20	23	17	19	22	16	19	21	18	21	26
Ni(II)	18	22	26	12	15	18	13	15	19	19	20	23	18	21	24	20	22	25
Cu(II)	22	25	32	13	15	19	16	18	21	15	18	20	20	23	25	18	21	25
Nystatin	-	-	-	-	-	-	-	-	-	20	22	24	23	25	27	20	23	29
Gentamycin	18	25	29	19	22	24	18	21	23	-	-	-	-	-	-	-	-	-

*Concentration in ppm
Zone of inhibition in mm

The bactericidal and fungicidal investigation data (Figures 5 and 6) of the compounds are summarized in Table 3. The results of the investigations account for the antipathogenic behavior of the compounds and this efficacy is positively modified on complexation. The biological activity the compounds against *E. coli* bacteria are found to follow the order Cu(II)-CDN>Gentamycin>Co(II)-CDN>Ni(II)-CDN>CDN (ligand). The order of increasing the biological activity the compounds against *S. aureus* bacteria are found to be Gentamycin>Co(II)-CDN>Cu(II)-CDN>Ni(II)-CDN>CDN (ligand). The order of increasing the biological activity the compounds against *S. fecalis* bacteria are found to be Gentamycin>Co(II)-CDN>Cu(II)-CDN>Ni(II)-CDN>CDN (ligand).

The biological activity the compounds against *A. niger* fungi follow the order Nystatin>Ni(II)-CDN>Co(II)-CDN>Cu(II)-CDN>CDN (ligand). The order of increasing the biological activity the compounds against *T. polysporum* fungi are found to be Nystatin>Cu(II)-CDN>Ni(II)-CDN>Co(II)-CDN>CDN (ligand). The order of increasing the biological activity the compounds against *C. albicans* fungi are found to be Nystatin>Co(II)-CDN>Ni(II)-CDN>Cu(II)-CDN>CDN (ligand).



Antibacterial activity of CDN Schiff base and its metal complexes against *E. coli*

Antibacterial activity of CDN Schiff base and its metal complexes against *S. aureus*

Antibacterial activity of CDN Schiff base and its metal complexes against *S. fecalis*

Antifungal activity of CDN Schiff base and its metal complexes against *A. niger*

Antifungal activity of CDN Schiff base and its metal complexes against *T. polysporum*

Antifungal activity of CDN Schiff base and its metal complexes against *C. albicans*

Figure 6: Antifungal screening data of CDN and its metal complexes**.**

4. CONCLUSIONS

In the present research studies, our efforts are synthesized of some newly compounds from the conventional as well as microwave methods. These synthesized compounds characterized by various physicochemical and spectral analyses. In the result of microwave assisted synthesis, it has been observed that the reaction time decreased from hours to minutes and availability of the product within better yields compared to the classical method. FAB-mass and thermal data show degradation pattern of the complexes. Thermogravimetric studied of the complexes also helped to characterize the complexes. The XRD patterns indicate crystalline nature of the complexes. The findings of the bactericidal and fungicidal investigation of the compounds against the opportunistic pathogens reveal that the synthesized compounds have the antipathogenic activity.

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