

ORIENTAL JOURNAL OF CHEMISTRY

An International Open Access, Peer Reviewed Research Journal

ISSN: 0970-020 X CODEN: OJCHEG 2024, Vol. 40, No.(5): Pg. 1449-1459

www.orientjchem.org

Synthesis, Physical, Spectral Characterization and Biological Studies of the complexes of Ni²⁺, Cu²⁺, Co²⁺ and Cd²⁺ ions with Schiff base Derived from p-hydroxybenzaldehyde & o-Phenyl-diamine

MOHAMMAD ABDUL ALIM¹, MD. ABUL BASHAR², PARTHA SARATHI ROY¹, MD. NURUZZAMAN KHAN³, PIJUSH KANTI ROY⁴, MD. SIDDIK ALI⁵, MD. REZAUL HAQUE ANSARY⁵ and MD. MOTAHAR HOSSAIN^{5*}

¹Department of Chemistry, Bangabandhu Sheikh Mujibur Rahman Science and Technology University, Gopalganj-8100, Bangladesh.

²Department of Biochemistry and Biotechnology, Khwaja Yunus Ali University, Sirajgonj-6751, Bangladesh. ³Department of Chemistry, Begum Rokeya University, Rangpur-5400, Bangladesh.

⁴Department of Chemistry, Mawlana Bhashani Science and Technology University, Santosh,

Tangail-1902, Bangladesh.

^{5*}Department of Chemistry, University of Rajshahi, Rajshahi-6205, Bangladesh. *Corresponding author E-mail: motaharbd7@gmail.com; motahar chem@ru.ac.bd

http://dx.doi.org/10.13005/ojc/400528

(Received: August 03, 2024; Accepted: September 05, 2024)

ABSTRACT

Together, ortho (o)-phenyldiamine and para (p)-hydroxy benzaldehyde generate a Schiff base. In order to make transition metal complexes of Ni2+, Cu2+, Co2+ and Cd2+ ions, this Schiff base was employed as a ligand of choice. The generated transition metal complexes' chemical structure is examined using a variety of physical methods, such as fundamental analysis, conductivity (molar), susceptibility (magnetic), spectroscopy (IR), and ultraviolet-visible spectroscopy. According to the elemental data analysis, a 1:2 [M:2L] complex of the formula, M^{2+} L₂ is produced (M^{2+} = Ni²⁺,Cu²⁺, Co²⁺, Cd²⁺ ions and L = Schiff base). All of the complexes were shown to be electrolytic in nature, as demonstrated by the molar conductance (conductivity) experiment. The ¹H NMR and infrared (IR) spectral studies were utilized to fix the Schiff base binding sites that the transition metal ions are attached to. The anticipated coordination geometry and magnetic characteristics, such as the magnet with paramagnetic or diamagentic of the complexes were validated by the magnetic susceptibility tests and electronic spectral data. While the Cd2+ ion creates tetrahedral structure with low spin, the Ni²⁺, Cu²⁺ and Co²⁺ ions yield high spin tetrahedra geometry. Comparing the obtained results with common antibiotics as kanamycin and ampicillin, the Cu2+ and Ni2+ complexes showed high activity, while the Co2+ and Cd2+ complexes showed week and occasionally moderate antimicrobial activity. The complex compounds of Schiff base showed more activity towards Gram-positive and Gram-negative bacteria as compared to its Schiff base. This idea can be improved upon with more adjustments and used in the pharmaceutical or medical industries.

Keywords: Schiff base, Metal complex, Spectral investigation and Anti-microbial action.

This is an <a>Open Access article licensed under a Creative Commons license: Attribution 4.0 International (CC- BY). Published by Oriental Scientific Publishing Company © 2018



INTRODUCTION

Schiff bases have a potential as well as a significant part in inorganic science as they can undoubtedly stable buildings with the majority of the transition metal ions¹⁻⁶. Specifically, Schiff bases are combined through the buildup of essential amine along with carbonyl compound under the particular condition7-12. Moreover, Schiff bases and its metal complex compound are likewise notable to show articulated organic action and structure a significant class of mixtures in medication and drug field²⁰⁻²⁷. Additionally, it demonstrated a degree of relaxing, antitumor, antibacterial, antifungal, and anticancer movement. The natural activity of Schiff base²⁸⁻³⁴ is attributed to the azomethine linkage. These ligands' transition metal complex molecules exhibit different configurations, underlying danger, and an aversion to atomic conditions. The central metal particles in these buildings go about as dynamic locales for pharmacological specialist³⁵⁻⁴⁶. This feature is utilized for displaying dynamic destinations in organic frameworks⁴⁷⁻⁵¹. Kudrat et al., carried out "synthesized, characterized evaluated mixed -ligand complexes of Cu2+ containing the Schiff base ligand derived from 2-hydroxybenxaldehyde with 2-amino phenol/3amino phenol and bidentate auxiliary ligands"35. The researchers noticed the buildings with Schiff bases showed the essential enemy of microbial action. p-hydroxybenzaldehyde can be extracted from Saprophytic perennial herb in the Orchidaceae family named orchids Gastrodia elata¹³ as well as found in Galeola faberi¹⁴. Chelating imines are available in Schiff bases, which have been broadly utilized as ligands to amalgamation of mesogenic complexes of Ni2+ and Cu²⁺ particles (ions)¹⁵. The synthesis of this kind of Schiff base complexes emphatically relies upon the idea of their substituents i.e., the presence and the shortfall of mesomorphism relies basically upon the chain length of the substituents. It was reported that "Some non-mesogenic tetradentate Schiff bases additionally give mesogenic compounds when complexing with copper"16-17. "The oxygen and nitrogen donor (O, N donor) ligands have a crucial role in the formation of transition metal coordination complexes"55-56. Adhao et. al. studied (i) "Synthesis, Spectral, Thermal Studies and Antimicrobial Evaluation of Transition Metal Complexes with Novel Schiff base Ligand⁵⁵," and (ii) "Synthesis, Structures and Antimicrobial activities of Novel Schiff base Ligand and Its Metal Complexe⁵⁶." Both the cases noted that transition metal complexes had increased antibacterial activitycompared to the Schiff base ligand.55-56. An enormous number of metal complex compounds with various electronic designs have been combined utilizing Schiff base ligands¹⁸⁻²⁰. At present, metal complexes of Schiff bases definitely stand out enough to be noticed because of their wonderful antibacterial, antifungal and antitumor exercises²⁶⁻³¹. Already, Saito et al., noticed a couple of transition metals buildings were incorporated and concentrated on their antimicrobial action³². It was seen that the designs (structure) have likely antimicrobial turn of events. Keeping these actual factors in mind-the significance of metal in physical, natural, climate, and clinical science, Schiff base was synthesized by union of para-hydroxybenzaldehyde and orthophenyldiamine and the complexation multiple transition metal particles (ions), including Ni²⁺, Cu²⁺, Co²⁺, and Cd²⁺ trasition metal cations through this Schiff base report here. Additionally, the complexes were categorized applying elemental examination, conductivity property and magnetic behavior along with IR (infrared), ¹H NMR spectra and UV-Vis (ultraviolet-vissible spectroscopy) in order to find out the geometrical shape of synthesized complexes. Moreover, the microbial activity of these building (complexes) have been in like manner proclaimed here. The aim of the work is to utilize this idea in medicinal or pharmaceutical industries for preparing antimicrobial drugs.

EXPERIMENTAL

Reagents (materials) and Chemicals

Unless otherwise specified, all additional reagents and solvent were analytical grade and utilized exactly as received. p-hydroxy benzaldehyde, o-phenyl diamine, absolute ethanol, and transition metal salts i.e., hydrated nickel nitrate, copper nitrate, cobalt chloride and cadmium nitrate were procured from Sigma–Aldrich chemicals. The ligands and its complexes were liquified in absolute alcohol.

Physical measurements

The melting points were measured for all metal complexes utilizing electro-thermal instrument for measuring melting point (Model no. AZ6512). Elemental analysis i.e., the percentage of elements (specifically Carbon, hydrogen & nitrogen) for ligands as well as complexes were executed using Organic Elemental Analyzer II (Model no. Perkin 2400) at the University of Kayama in Japan. The measurement for magnetic moments were taken by utilizing Susceptibility Balance (Magnetic) made by The SHERWOOD SCIENTIFIC. Pascal's constant was used to do diamagnetic adjustments³³. FT-IR spectra for the ligand & its complexes of different transition metals were recorded through utilizing FTIR spectrophotometer (Model no. NICOLET 310) made from Belgium in the frequency ranges from 4000 to 400 cm⁻¹. This experiment was done in the Division of Metallurgy and Material Science, BUET where, KBr was used as disc for FT-IR spectro-photometer. TMS was utilized as internal standard, the spectra of ¹H NMR (300MHz) were captured using a NMR (Model no. JEOL JN M-A400) spectrometer in DMSO. UV-Visible spectral information was taken utilizing a spectrophotometer named SHIMADZU DOUBLE BEAM (model UV-1200) in the range of wave length of 200-900nm. This experiment was carried out at Rajshahi University's Central Science Laboratory in Bangladesh.

Schiff base synthesis

The synthesis of Schiff base was carried out through continuous stirring of the mixture of ethanoic solution of para (p)-hydroxy benzaldehyde and ortho (o)-phenyldiamine. In order to prepare the mixture, 10 mmol ethanolic solution of o-phenyldiamine (1.08 g o-phenyldiamine in 20 mL in absolute ethanol) was mixed with 10 mmol ethanoic solution of p-hydroxy benzaldehyde (1.22 g p-hydroxy benzaldehyde in 30 mL absolute ethanol).

At standard (room) temperature, the mixture was assorted and stirred for four hours before being allowed to stand for twenty-four hours. A yellow crystal was formed. A Buchner funnel was used to have the yellow crystal, which was then filtered out and desiccated using vacuum desiccator containing CaCl₂ (anhydrous). The following illustrates the reaction flow chart (Scheme 1) to explain the synthesis of Schiff base.



The initial step is an elongation of a nucleophillic to the =C=O group in the imine development system (Scheme 2). For this purpose, the amine acts as nucleophile, responds with the ketone or aldehyde or produce carbinolamine, an imbalanced expansion product. As shown in Scheme 2, a carbinolamine is a chemical species linked to an amine bunch ((-NH₂)-NHR or -NR) and a hydroxy radical assembling to a comparable carbon.



Scheme 2. Reaction mechanism scheme for synthesizing Schiff base

A reasonable amount of production found to be 60-65% was gained and elemental analysis observation confirmed the complex compound's purity.

Complexes synthesis with Schiff base

One millimol of hydrated nickel nitrate or copper nitrate or cobalt chloride or cadmium nitrate was liquified separately with 10 milliliters pure ethyl alcohol. Each and every salt solution received ethanoic solution of two mmol of the Schiff base, which was continuously agitated for four hours at room temperature before being left to remain still for one hour. Filtered off, a vacuum desiccator with anhydrous CaCl₂ was used to dry the residue. Different colored solid metal complexes precipitated out after reaching normal temperature. The obtained products underwent filtering, a cold methanol wash, and drying. Below are the general reaction scheme and the suggested structure (Fig. 1) of the produced complexes. $\mathsf{M}(\mathsf{NO}_3)_2.\mathsf{xH}_2\mathsf{O} + 2\mathsf{SB} \rightarrow [\mathsf{M}(\mathsf{SB})_2]^{2+}. (\mathsf{NO}_3)_2$

 $\label{eq:Where, M2+} Where, M^{2+} = Ni^{2+}, Cu^{2+}, Co^{2+} \mbox{ and } Cd^{2+} \mbox{ ions}, \\ SB = Schiff \mbox{ base }$



Fig. 1. The predicted tetrahedral geometry of complexes

Antimicrobial Screening of the metal complexes There exist several *in vitro* techniques for determining an organism's susceptibility to an antimicrobial agent. The disc diffusion technique is generally accepted for use in initial studies of substances suspected of having antibacterial properties^{1,2}. The complexes' antimicrobial activities of Ni²⁺, Cu²⁺, Co²⁺ and Cd²⁺ ions was carried out by the disk diffusion technique⁵². This analysis was done on four pathogenic bacteria i.e., two-gram negative bacteria like *Escherichia coli*, *Shigella dysenteriae* and *Gram-positive* bacteria like *Bacillus cereus* and *Streptococcus agalactiae* at 37°C. A disk containing filter paper (Whatmann No. 4) with diameter (8mm) was immersed in a DMF compound solution (1.0 mg cm⁻¹)⁵². It was put on nutritional agar plates after drying. After 36 h, the inhibitory zones were seen. Ampicillin and kanamycin were utilized as standards, and DMF served as a control.

RESULTS AND DISCUSSION

At normal (room) temperature, all complexes are stable. While none of the complexes are soluble in conventional organic solvents, they are all very soluble in DMSO, DMF, and CHCl_a.

Elemental examination and conductivity measurement

Tables 1 and 2 present the physical parameters of the produced complex compounds and the findings of the conductivity investigations. For the complex compounds, the majority of the analytical findings agreed through the anticipated empirical recipe. The conductance value for the complex compounds uncovered that the nature of these species (complexes) are electrolytic²⁸.

Ligand (SB) and complexes % C % H % N Computed Observed Computed Observed Computed Observed SB*(C₁₃H₁₃N₂O) Yellow crystal 73.58 73.25 5.53 5.60 13.20 13.12 [Ni(C₁₃H₁₂N₂O)₂]²⁺ 64.64 64.02 4.97 4.32 11.60 10.98 $[Cu(C_{13}H_{12}N_{2}O)_{2}]^{2+}$ 64.12 63.30 4.92 4.16 11.48 11.02 [Co(C₁₃H₁₂N₂O)₂]²⁺ 64.60 64.15 4.96 4.28 11.59 11.16 $[Cd(C_{13}H_{12}N_{2}O)_{2}]^{2+}$ 57.42 57.02 4.42 5.86 10.30 9.78

Table 1: The ligand's and complex's fundamental analysis

Table 2: Physical analysis of Schiff base (ligand) & their complex compounds

Complex compounds	Colour	Melting point or decomposition temp.($\pm 5^{\circ}C$)	%Yield	Molar conductance(ohm ⁻² cm ² mol ⁻¹)
[Ni(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	Yellowish	255(d)	62	98
[Cu(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	Black	280(above)	65	115
[Co(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	Black	235(d)	61	121
[Cd(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	White	270(above)	60	120
SB(C ₁₃ H ₁₂ N ₂ O)	Yellow crystal	150	-	-

*Schiff base

Infrared (IR) spectral studies

Table 3 lists the vibrational frequencies for transition metal complexes along with their approximative assignments. Comparing with the frequency of vibration of the free ligands & related compounds made the assignments easier. Table 3 contains infrared (IR) spectrum data for ligand and its complexes. The ligand's spectral data revealed regions of strength for a band at 1550-1651 cm⁻¹ caused by v(C=N) stretching, signifying that a buildup has taken place between the ortho (o)-phenyldiamine²⁹⁻³⁰ and the CHO group of para-hydroxy benzaldehyde. The infrared spectra of complexes have two conceptual characteristics. The azomethine group's (=C=N-) stretching frequencies of the transition metal complexes have shifted to downward and are located in the span of 1574-1610 cm⁻¹ in contrast to the free ligand bands at 1613 cm⁻¹. This shift may be caused through the coordination of metal ions with two azomethine groups. The second characteristic is found in the band or frequency at (500-600) cm⁻¹ due to (M-N) stretching³¹⁻³², which showed the metal ions were definitely coordinated with the produced Schiff base

via the N atom. This band was not seen for the free Schiff base, confirming that the metal ion and Schiff base had formed an M-N bond in all of the complex compounds. The band that was found falls between 3440 and 3510 cm⁻¹ and can be characterized to the vibrating frequencies of the Schiff base's (O-H) bond, which is not correlated (coordinated) with the metal ions. A band at (3045-3165) cm⁻¹ range was seen as a result of the Schiff base's aromatic (C-H) vibration. A band within the span of (3300-3500) cm^{-1} was discovered for the stretching of $v(NH_2)$ modes. A strong value was ascribed to the noncoordinate nitrate in each complex, and this value is supported by the conductivity measurement readings for each complex. The values vary from 1383 to 1885 cm⁻¹.

Table 3: IR (infra-red spectra) information of the ligand & its metal complexes (cm⁻¹)

SB and metal complexes	ν (O–H)	ν (NH ₂)	ν (C=N) of aromatic ring	ν (C–H)	ν (M–N)
$[Ni(C_{13}H_{12}N_2O)_2]^{2+}$	3435.64	3318.46	1607.94	3126 3205 14	578.39
$[Co(C_{13}H_{12}N_{2}O)_{2}]^{2+}$ $[Co(C_{13}H_{12}N_{2}O)_{2}]^{2+}$	3551.30	3411.64	1609.55	3056.84	515.75
$\frac{[Cd(C_{13}H_{12}N_2O)_2]^{2^+}}{SB^*(C_{13}H_{12}N_2O)}$	3421.54 3402.29	3303.10 3318.33	1629.73	3042.99 3010.37	523.98

*Schiff base

Figures 2a, 2b, 2c, 2d, and 2e demonstrate spectra of FT-infrared for the Schiff base and its





Fig. 2(c). FTIR spectra for Cu²⁺ complex with Schiff base

combinations with transition metal ions of Ni²⁺, Cu²⁺, Co²⁺ and Cd²⁺ are displayed as below:



4000.0 3600 3200 2800 2400 2000 1800 1600 1400 1200 1000 800 600 400 225.0 cm⁻¹





Fig. 2(d). FTIR spectra for Co2+ with Schiff base



Fig.1H NMR spectra analysis

Ligand's structure was confirmed by applying the ¹H NMR spectral data. The ligand (SB)'s ¹H NMR spectra, which were recorded in DMSO solvent (Fig. 3), revealed well-resolved signals for the ligand's protons. The compounds' ¹H NMR spectra were obtained in the identical circumstances. For the aromatic protons, the spectra showed a multiplet at δ 6.50-7.65ppm. In the case of the Schiff base, the imine (-CH=N-) and (-NH₂) protons displayed two singlets at δ 8.48 and 5.24ppm, respectively, which shifted to δ 8.01 and 5.08ppm for the complex. This implies that the nitrogen (-NH₂₎ and azomethine (-CH=N-) have been coordinated to the metal center. Additionally, a singlet was detected at δ 9.54ppm for the ligand's phenolic proton, which does not alter upon complexation.





Magnetic Moment and Electronic Spectra

Magnetic moment values of the produced compounds at normal condition are shown in Table 4. From the data, it is observed that the mass susceptibility values for Ni²⁺, Cu²⁺ and Co²⁺ are positive but for Cd²⁺ is negative. Table 4 also lists the outcomes of the calculation of the magnetic moment for each metal complex using mass susceptibility data. Regarding two (2) unpaired electrons, 3.27 B.M. was discovered to be the magnetic moment value for Ni²⁺ complex. This value demonstrates paramagnetic and high spin tetrahedral geometry of Ni²⁺ complex. The values of 2.45 B.M. and 3.99 B.M. as magnetic moment for Cu2+ and Co2+ complexes, respectively, confirm one and three unpaired electrons. Therefore, Cu2+ and Co2+ complexes are both high spin tetrahedral complexes with one and three unpaired electrons, respectively, as indicated by the magnetic moment data. The lack of magnetic moment was attributed to the Cd²⁺ complex's mass susceptibility having a negative value. The Cd2+ complex's diamagnetic magnetic moment worth indicates the lack of unpaired electrons, as indicated by the negative mass susceptibility value. Consequently, it can be said that Cd²⁺ has a diamagnetic character. The attractive second information indicates that the Cd²⁺ combination is a tetrahedral complex with a low spin³³.

The electronic spectra for all complexes show a broad range of bands because of -CH=N-. The result of the metal and ligand's electron cooperation is charge movement, which can include both either ligand-to-metal and metal-to-ligand electron movement⁵³. The bands seen in the 235-265nm region are caused by the C=N group⁵⁴ due to $\pi \rightarrow \pi^*$ transition. Because nitrogen participated in coordination with the metal ion, the band was moved up to a more elevated range.

The absorption band is visible in the 310–370nm region because of the $n \rightarrow \pi^*$ transition from the imine group that corresponds to the metal complexes or ligands. The compounds' electronic spectrum data are displayed in Table 5. DMF solvent was used in order to evaluate the electronic spectra for all the produced complexes. In the complexes of Ni²⁺, Cu²⁺, Co²⁺ and Cd²⁺ ions, the electronic spectra bands at 310nm, 270nm, 260nm and 308nm were observed, respectively. These bands of the compounds were observed in the region of 200-420nm. These bonds were only reflected due to charge transfer in the specified range (200–420nm)³⁴.

Table 4: Analytical data for magnetic moment of the complexes

No	Complexes	Length of the sample 'I' in cm	Weight of the sample 'm' in g	Susceptibility of the emply tube 'R _o '	Susceptibility of the sample with tube 'R	Mass susceptibility X _g ×10 ⁻⁶ CGS Unit	Molar weight of the sample, N	Molar susceptibility X _g ×10 ⁻⁶ 1 CGS Unit	Diamagnetic correction X _L ×10 ⁻⁶ CGS Unit	c χm _{corr} ×10 ⁻⁶ C.G.S. unit	μ _{eff} in Β. Μ.
1	[Ni(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁻	+ 2.1	0.0252	-63	-16	8.710	482.69	4204.22	-257.40	4461.62	3.27
2	[Cu(C ₁₃ H ₁₂ N ₂ O) ₂] ²	+ 2.2	0.0278	-55	-27	4.622	487.50	2253.27	-255.40	2508.62	2.45
3	[Co(C ₁₃ H ₁₂ N ₂ O) ₂] ²	+ 2.7	0.0300	-65	+11	13.91	482.93	6379.50	-257.40	6636.90	3.99
4	$[Cd(C_{13}H_{12}N_{2}O)_{2}]^{2}$	+ 2.0	0.0228	-53	-58	-0.724	536.41	-388.36	-259.40	-128.96	Dia.

Table 5: The results of electronic spectral for metal complexes

No	Complexes	λ _{max} (nm)
1	[Ni(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	310
2	[Cu(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	270
3	[Co(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	250
4	[Cd(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	308

Antibacterial screening of the metal complexes: Estimating the zone of hindrance assessed surrounding the location, as shown in Table 6, allows for the communication of antimicrobial exercises of the test samples (Schiff base & their metal complexes). The results showed that the metal complex compounds are more toxic to microorganisms than either ligands or free metal particles (ions). The metal complexes were assessed towards the four pathogenic bacteria were tested and it was observed that complexes categorically exhibited moderate to low levels of activity.

Table 6: Antimicrobial activity for Schiff base, metal complexes & standard kanamycin

Compounds				
	E. coli	S. dysenteries	B. cereus	S. agalactiae
Schiff base	Nill	Nil	Nil	Nil
[Ni(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	8	15	15	12
[Cu(C ₁₃ H ₁₂ N ₂ O) ₂] ₂₊	19	9	7	15
[Co(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	6	11	5	9
[Cd(C ₁₃ H ₁₂ N ₂ O) ₂] ²⁺	9	-	6	7
Kanamycin ³⁰	32	26	32	31
DMSO (Control disc)	Nil	Nil	Nil	Nil

The results also revealed that these metal structures, or complexes, exhibited strong resistance to both Gram-positive and Gram-negative microscopic organisms (bacteria) in all of the instances that were studied. The graphical representation of the activity for the standard Kanamycin, Schiff base and its complexes towards two Gram-negative (E. coli, S. dysenteriae) and two Gram-negative (B. cereus and S. agalactiae) bacteria are demonstrated below (Fig. 4). The Ni²⁺ as well as Cu²⁺ complex compounds exhibited the good performance against above four pathogenic bacteria among the four complexes. Complex Cd2+ showed zero activity against Streptococcus agalactiae whereas it exhibited less activity for the remaining three bacteria.

Conversely, Cu2+ compound demonstrated

more potency (highest activity) compared to other metal complexes against Escherichia coli. Co2+ complex compound showed significant activity against all bacteria (pathogenic). All the outcomes are related with the standard antibiotics like, ampicillin and kanamycin. Ampicillin and Kanamycin both showed almost the same activity against four pathogenic bacteria. Therefore, analytical result for kanamycin is only mentioned in the above Fig. 2. The activity showed for the metal complexes are smaller compared with standard Ampicillin and Kanamycin as noticed in the Table 6. From table, it is clearly observed that the synthesized Schiff base did not display a little activity against all four bacteria. Therefore, it is also crystal clear that the metal ions in the complex compounds are responsible to play the key role to show good activities, because the ligand did not show any activity against any pathogenic bacteria.



Fig. 4. Ni²⁺, Cu²⁺, Co²⁺ and Cd²⁺ metal ion complexes & Schiff base's antimicrobial activities against four pathogenic bacteria. Kanamycin was used as standard

CONCLUSION

Magnetic susceptibility data, or magnetic moment values, confirm that the Cd²⁺ complex with Schiff base is diamagnetic in nature, whereas the Ni²⁺, Cu²⁺ and Co²⁺ complexes with the same one are paramagnetic. Every single metal particle (i.e., ion) was revealed to be coordinated by two Schiff base's N (N of -N=CH- and N of amine group of Schiff base) atoms. The values of magnetic moment for Cd2+ complex reveal tetrahedral structure with low spin form, while other three metal ions Ni2+, Cu2+ and Co²⁺ with Schiff base have high spin along with tetrahedral structure. Additionally, the electronic spectrum data confirmed that each compound has a tetrahedral structure. Each and every compound was found to be ionic in nature based on the molar conductivity values. Stretching frequencies (IR data) for uncoordinated nitrate ions confirm that the nature of the entire complexes are ionic, which is also affirmed by the molar conductivity values. It is concluded that for every complex, the free nitrate ion is situated outside the coordination sphere. As demonstrated by standard Kanamycin and Ampicillin, the Cu2+ complex exhibits the most notable (extreme strong) antibacterial effect towards Gram-positive and Gram-negative harmful germs

- Hamil, A. M.; Khalifa, K. M.; Al-Houni, A.; El-Ajaily, M. M.; Synthesis, spectroscopic investigation and antiactivity activityof Schif base complexes of cobalt(II) and copper(II) ions., *Rasayan Journal of Chemistry.*, 2009, 2, 261–266.
- Mishra, A. P.; Sharma, N.; Synthesis, characterization, X-ray and thermal studies of some Schif base metal complexes., *J of the*

(microorganism). Antibacterial movement of Schiffbased Ni²⁺, Co²⁺ and Cd²⁺ structures (complexes) against examined harmful microscopic organisms is moderate to low.

The nitrogen of amine group and of the azomethine group were discovered to coordinate the ligand (SB), generating stable chelates with a metal to ligand ratio of 1:2. It was demonstrated that at room temperature, the metal complexes remained stable. The results of all the aforementioned studies and examinations pointed to the creation of stable metal complexes by the selected metal (II) ions, amine, and nitrogen donor Schiff base ligand. It was also shown that the complexes of Cu^{2+} and Zn^{2+} ions exhibited more enhanced antibacterial activities than the ligand.

ACKNOWLEDGEMENT

The writers of this work may wish to acknowledge the financial assistance provided by Bangladesh's Science and Technology Ministry, Bangladesh. Likewise, the chairperson of the Chemistry Department at the University of Rajshahi, Bangladesh, and the Central Science Research Laboratory, Rajshahi University deserves recognition, for providing the required instrumental support and readily available chemicals. The professor of the Organic Synthesis Laboratory at Kayama University's Faculty of Science in Japan is also much appreciated for his examination of the complexes' elemental analysis.

Conflicts of interest

Every co-author has reviewed the work and confirmed its accuracy; none of them has any conflicts of interest to disclose. We attest that the submission is entirely unique and isn't being considered for publishing with any other works.

REFERENCES

Indian Council of Chem., 2009, 26, 125–129.

- Crian, R.; Modra, D.; The synthesis of salicylaldehyde varying different parameters.
 J. West University of Timi oara, Series of Chemistry., 2013, 22, 57-66.
- Simmonds, J.; Robinson, G.K.; Formation of Benzaldehyde by Pseudomonas putida ATCC 12633., *Appl. Microbial Biotechnol.*, **1998**, *50*, 353-358.

- Dueke-Eze, C. U.; Fasina, T. M.; Idika, N.; Synthesis, electronic spectra and inhibitory study of some Salicylaldehyde Schiff bases of 2-aminopyridine., *African Journal of Pure and Applied Chemistry.*, **2011**, *5*, 13-18.
- Schiff, W.; Szady, C. A.; Grech, E.; Przybylski, P.; Brzezinski, B., Spectroscopic studies of new Schiff and Schiff–Mannich bases of ortho-derivatives of 4-bromophenol., *J. Mol. Struct.*, 2002, *643*, 115-121.
- Schiff H., Mittheilungen aus dem Universitätslaboratorium in Pisa: Eine neue Reihe organischer Basen., *Justus Liebigs Ann Chem.*, **1864**, *131*(1), 118–119.
- Bell, S. S. C.; Conklin, G. L.; Childress, S. J.; The Separation of Ketimine Isomers., *J. Am. Chem. Soc.*, **1963**, *85*, 2868–2869.
- Prisakar, V. I.; Tsapkov, V. I.; Buracheeva, S. A.; Byrke MS, Gulya AP Synthesis and antimicrobial activity of coordination compounds of copper with substituted salicylaldehyde thiosemicarbazones., *Pharm. Chem. J.*, **2005**, *39*, 30-32.
- Naturforsch, Z.; Pelttari, E.; Karhumaki, E.; Langshaw, J.; Perakyla, H.; Elo, H.; Antimicrobial properties of substituted salicylaldehyde and related compounds., *J. Biosci.*, 2007, 62C, 487-497.
- Tsapkov, V. I.; Prisacar V. I., Buracheva, S. A.; Lazakovich, D.V.; Gulya, A.P; Synthesis and antimicrobial activity of sulfazine-containing copper(II) coordination compounds with substituted salicylaldehyde benzoyl hydrazones., *Pharm. Chem. J.*, **2008**, *42*, 523-526.
- 12. Shi, L.; Ge, H. M.; Tan, S. H.; Li, H. Q.; Song, Y. C.; Zhu, H. L.; Tan, R. X.; Synthesis and antimicrobial activity of Schiff bases derived from 5-chloro-salicylaldehyde., *Eur. J. Med. Chem.*, **2007**, *42*, 558-564.
- Ha, J.H.;Lee, J.T.;Kim, J.S.;Yong, C.S.;Kim, J.A.; Ha, J. S.; Huh, K.; 4-Hydroxybenzaldehyde from Gastrodia elata B1. is active in the antioxidation and GABAergic neuromodulation of the rat brain., *Journal of Ethnopharmacology.*, **2000**, *73* (1-2), 329-333.
- Li, Y. M.; Zhou, Z. L.; Hong, Y. F.; Studies on the phenolic derivatives from Galeola faberi Rolfe., *Yao xue xue bao=Acta pharmaceutica Sinica (in Chinese).*, **1993**, *28*(10), 766-771.
- 15. Iskander, M. F.; El Sayed L.; Coordination

compounds of hydrazine derivatives with transition metals-IV Ni (II)-complexes with hydrazinedithiocarboxylic acid, its methyl ester and their N-isopropylidine derivatives., *J. Inorg. Nucl. Chem.*, **1971**, *33*, 4253-4263.

- 16. R. Pasche, H. Zaschke, A. Madicke, J. R. Chipperfield, A. B. Blake, P. G. Nelson and G. W., *Gray, Mol. Cryst. Liq. Cryst Lett.*, **1988**, *6*, 81,
- Shaffer, T. D.; Sheth, K. S.; Mesomorphic Transition Metal N₂O₂ Chelates., *Mol. Cryst. Liq. Cryst.*, **1989**, *172*, 27-29.
- Foye, W. O.; Duvall, R. N.; Metal chelates and antitubercular activity. III. p-Aminosalicylic acid: chelate vs. complex., *J. Am. pharm. Accoc. Sci. Educt.*, **1958**, *47*, 282-285.
- Dilworth, J. R.; The coordination chemistry of substituted hydrazines., *Coord. Chem Rev.*, 1976, 21(1), 29-62.
- Kayal, M.; Dutt, Y.; Analytical applications of hydrazones., *Talanta.*, **1975**, *22*(2), 151-166,
- E-Zahan, M.K; Islam, M.S.; and Bashar M.A.; Synthesis, Characterization and antimicrobial activity of Some Metal Complexes of Mn(II), Fe(III) Co(II), Ni(II), Cu(II) and Sb(III) Containing Bidentate Schiff base of SMDTC., *Russian Journal of general chemistry.*, **2015**, *85*(3), 667-672.
- Uddin, M. N.; Chowdhury, D. A.; Rony, M. M.; Halim, M. E.; Metal complexes of Schiff bases derived from 2-thiophenecarboxaldehyde and mono/diamine as the antibacterial agents., *J. Science publishing group.*, **2014**, *2*, 6-14.
- 23. Jamuna, K.; Naik, B. R.; Sreenu, B.; Seshaiah, K.; Synthesis, characterization and antibacterial activity of Cu(II) and Fe(III) complexes of a new tridentate Schiff base ligand., *J. Chem. Pharm. Res.*, **2012**, *4*, 4275-4282.
- Uddin, M. M.; Salam, M. A.; Sultana, J.; Pb(II) complexes of Schiff bases derived from benzoylhydrazine as the antibacterial agents., *J. Science publishing group.*, 2015, *3*, 7-14.
- 25. El-Ajaily, M. M.; Abdlseed, F. A.; Gweirif, B.; Preparation, characterization and antibacterial activity of some metal ion complexes., *E-Journal of Chem.*, **2007**, *4*, 461-466.
- Sharif, S. A. I.; El-Tajoury, A. N.; Elamar, A. A.; Preparation and Antibacterial Activity of Mixed Ligand Complexes of Co(II), Ni(II), Cu(II) and Cd(II) Derived from 1-Phenylazo-2-naphthol and Salicylaldehyde., *E-Journal of Chemistry.*, 2011, *8*, 43-48.

- Ran X.; Wang, L.; Cao, D.; Lin, Y.; Hao, J.; Synthesis, characterization and in vitro biological activity of cobalt(II), copper(II) and zinc(II) Schiff base complexes derived from salicylaldehyde and D, L-selenomethionine., *Appl. Organometal. Chem.*, **2011**, *25*, 9–15.
- 28. Geary, W.J., The Use of Conductivity Measurements in Organic Solvents for the Characterisation of Coordination Compounds., *Coord. Chem. Rev.*, **1971**, *7*, 81-122.
- Ashrafuzzaman, M.; Camellia, F.K.; Rahman, M. H.; Hossain M. D.; Hossain, M. T.; Shovon, M. G. M.; Paul, G. K.; Md. Zahan, K.E.; Haque, M. M.; Synthesis, Characterization, Antibacterial and Antioxidant Activities of Cu(II), Ni(II), and Zn(II) Mixed Ligand Complexes Containing Isoniazid Based Bidentate Schiff Base., Asian Journal of Chemical Sciences., 2023, 13(5), 112-122.
- Sadler, P. J.; Nast, M.; Narayaran, L. L.; The design of metal complex as anticancer drugs., Murtius-Nijhoff, Boston., **1989**.
- Hutchinson, B.; Takemoto, J.; and Nakamoto, K.; Metal isotope effect on metal-ligand vibration. II. Tris complexes of 2, 2/-bipyridine and 1, 10-phenanthroline., J. Am. Chem. Soc., 1970, 92, 3335-3339.
- Saito, Y.; Takemoto, J.; Hutchinson, B.; Nakamoto, K.; Infrared studies of coordination compounds containing low-oxidation state metals. I. Tris (2,-2/-bipyridine) and tris (1, 10-phenanthroline) complexes., *Inorg. Chem.*, **1972**, *11*(9), 2003-2011.
- Figgis, B.N.; Lewis, J.; The magnetic properties of transition metal complexes., *Prog. Inorg. Chem.*, **1964**, *6*, 73.
- 34. Lever A.B.P.; Inorganic Electronic Spectroscopy., **1984**, 2nd Edn. Elsevier.
- Shampa, J. A.; Islam, M. R.; Hossain, M. S.; Rahman, G. T.; Zakaria, C. M.; Zahan, M. K.; Physiochemical and Antibacterial Activity Investigation on Noble Schiff Base Cu(II) Complex., *American Journal of Heterocyclic Chemistry.*, **2017**, *3*(4), 37-41.
- Patel, K. M.; Patel K. N.; Patel, N. H.; Patel, M. N.; Synthesis, characterization, and antimicrobial activities of some transition metal complexes with a tridentate dibasic Schiff base and bidentate 2, 2'-bipyridylamine. Synth., *React. Inorg. Met-Org. Chem.*, 2001, 31, 239-246.

- Temel, H.; Cakir, Ü.; Ugras, H. I.; Sekerci, M.; The Synthesis, Characterization and Conductance Studies of New Co(III), Cu(II), Ni(II) and Zn(II) Complexes with Schiff Base Derived from 1,2-bis-(o-aminophenoxy) Ethane and Salicylaldehyde., *J. Coord. Chem.*, **2003**, 56, 943-951.
- Liang, Y.; S. U. B.; Zhao, I.; Sun, W.; The Synthesis of New Asymmetric Double Schiff Bases Containing a New o Amino Benzoic Acid Derivative., *Synthesis Communications.*, 2004, *34*, 3235-3242.
- Venkataraman, N. S.; Kuppuraj, G.; Rajajopal, S.; Metal-salen complexes as efficient catalysts for the oxygenation of heteroatom containing organic compounds-synthetic and mechanistic aspects., *Coord. Chem. Rev.*, 2005, 249, 1249-1268.
- Mohamed G. G.; Omar, M. M.; Hindy, A. M.; Metal complexes of Schiff bases: preparation, characterization and biological activity., *J. Turk. Chem.*, **2006**, *30*, 361-382.
- Diego, M.; Veronica, D.; Claudia, P.; Jean, P.T.; Sandra, S.; Synthesis, characterization and antioxidant activity of water-soluble Mn (III) complexes of sulphonato-substituted Schiff base ligands., *J. Inorg. Biochem.*, **2010**, *104*, 496-502.
- Khalil, M.; Ismail, E.; Mohamed, G., Zayed, E.; Badr, A.; Synthesis and characterization of a novel schiff base metal complexes and their application in determination of iron in different types of natural water., *Open J. Inorg. Chem.*, **2012**, *2*, 13-21.
- Abu-El-Wafa, S. M.; El-Wakiel, N.A.; Issa, R.M., Mansour, R.A.; Formation of novel mono-and multi-nuclear complexes of Mn(II), Co(II) and Cu(II) with bisazo-dianils containing the pyrimidine moiety: Thermal, magnetic and spectral studies., *J. Coord. Chem.*, **2005**, *58*, 683-694.
- Etaiw, S. H.; Abd El-Aziz, D. M.; Abd El-Zaher, E. H.; Ali E. A.; Synthesis, spectral, antimicrobial and antitumor assessment of Schiff base derived from 2-aminobenzothiazole and its transition metal complexes., *Spectrochim. Acta* A., **2011**, *79*, 1331–1337.
- 45. Ueda, J. I.; Takai, N.; Shiazue, Y.; Reactive Oxygen Species Generated from the Reaction of Copper(II) Complexes with Biological Reductants Cause DNA Strand Scission., *Arch. Biochem. Biophys.*, **1998**, *357*, 231-239.

- Radhakrishnan, P.K., Complexes of lanthanide perchlorates with 4-N-(2'-hydroxy-1'naphthylidene) aminoantipyrine., *Polyhedron.*, 1986, *5*, 995-998.
- Ashrafuzzaman, M.; Camellia, F. K.; Hossain, M. D.; Shovon, M. G. M.; Rahman, H.; Hossain, M. T.; Rahman, M. H.; Zahan, M. K-E.; and Haque, M. M.; Mixed Ligand Complexes of Ni²⁺, Cu²⁺ and Zn²⁺ Ions Containing N-(4methoxybenzylidene)isonicotinohydrazone Schiff Base and 1,10-Phenanthroline: Synthesis, Characterization, Antimicrobial and Antioxidant Properties., *Asian Journal* of Chemical Sciences., **2023**, *13*(6), 46-57.
- Islam, T.; Bitu, N. A.; Hossen, F.; Asraf, A.; Zahan, K-E.; Investigation on Biological Activities of Thiosemicarbazide Derived Schiff Base-Metal Complexes., *Asian J. Research Chem.*, **2024**, *17*(1), 6-12.
- Hossain, M. S.; Khushy, K. A.; Latif, M. A.; Hossen, M. F.; Asraf, M. A.; Zahan, M. K-E.; Abdou, A.; Co(II), Ni(II), and Cu(II) Complexes Containing Isatin-Based Schiff Base Ligand: Synthesis, Physicochemical Characterization, DFT Calculations, Antibacterial Activity, and Molecular Docking Analysis., *Russian Journal of General Chemistry.*, **2022**, *92*(12), 2723–2733.
- Camellia, F. K.; Ashrafuzzaman,,M.; Banu, L. A.; Islam M. N.; Md. Zahan, K-E.; Synthesis, Characterization, Antibacterial and Antioxidant Studies of Isoniazid-based Schiff Base Ligands and Their Metal Complexes., *Asian Journal of Applied Chemistry Research.*, 2022, 11(3), 8-23,
- 51. Bitu, M. N. A.; Hossain, M. S.; Asraf, M. A.; Hossen, M..F.; Zahan, M-E.; Mixed

Ligand Oxovanadium(IV) Complexes of 5,6-Dimethyl-1,10-phenanthroline and Amino acid Schiff bases Containing Salicylaldehyde: Synthesis and Characterization with Antibacterial Properties., *Afr. J. Pharm. Sci.*, **2021**, *1*(1), 47-53.

- Akila, E.; Usharani, M.; Ramachandran, S.; Jayaseelan, P., Velraj, G., Rajavel, R.; Tetradentate-arm Schiff base derived from the condensation reaction of 3,30 dihydroxybenzidine, glyoxal/diacetyl and 2-aminophenol: Designing, structural elucidation and properties of their binuclear metal(II) complexes., *Arabian Journal of Chemistry.*, 2017, 10, S2950–S2960.
- Tas, E.; Aslanoglu, M.; Kilic, A.; Kara, Z.; Synthesis, spectroscopic and electrochemical studies of copper(II) and cobalt(II) complexes of three unsymmetrical vic-dioximes ligands., *J. Coord. Chem.*, **2006**, *59*(8), 861–872.
- Serbest, K.; Karabocek, S.; Degirmencioglu, I.; Guner, S.; Kormali, F.; Mono, di- and trinuclear copper(II) dioxime complexes; 3-{2-[2-(2-hydroxyimino-1 methylpropylidene amino) ethylamino]ethylimino} butan-2-one oxime., *Trans. Met. Chem.*, 2001, *26*, 375-379.
- 55. Adhao, S.T.; Wagh, R. R.; Synthesis, Spectral, Thermal Studies and Antimicrobial Evaluation of Transition Metal Complexes with Novel Schiff base Ligand., *Orient. J. Chem.*, **2024**, *40*(1), 142-151.
- Adhao, S. T.; Wagh, R. R.; Synthesis, Structures and Antimicrobial Activities of Novel Schiff Base Ligand and Its Metal Complexes., *Orient. J. Chem.*, **2023**, *39*(6), 1571-1578.