

2,3- DIMETHOXYBENZALDEHYDE SCHIFF BASE OF SBDTC AND ITS METAL COMPLEXES OF Ni(II), Cu(II), Zn(II) AND Cd(II): SYNTHESIS, CHARACTERIZATION AND BIOLOGICAL ACTIVITY

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Abstract. A Schiff base containing Nitrogen-Sulphur donor, benzyl 2-(2,3-dimethoxybenzylidene)hydrazinecarbodithioate, was derived from the condensation of 2,3-dimethoxybenzaldehyde with S-benzylidithiocarbamate. The Schiff base and its complexes with Ni(II), Cu(II), Zn(II) and Cd(II) were synthesized and characterized by ¹H NMR, FT-IR, ESI-MS, elemental analysis, electronic spectra and various physico-chemical techniques. In all the complexes, the Schiff bases behaved as bidentate, uninegative ligands, giving square planar geometrical structures for Ni(II), Cu(II) and tetrahedral for Zn(II) and Cd(II) complexes. The Schiff bases and its complexes were screened for antibacterial activities towards potential antimicrobial application. The biological activity results revealed that the complexes were more microbial toxic than the free ligand. In particular, Cu(II), Ni(II) and Cd(II) complexes were found to be more effective antibacterial agents with respect to the Zn(II), which was found moderately active against the same bacteria.

Keywords: Metal complexes, SBDTC, Schiff base, biological activity.

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1. Introduction

The study of nitrogen-sulphur donor ligands like Dithiocarbamate, $\text{NH}_2\text{NHCS}_2^-$ and its substituted derivatives continues to be of great interest to researchers because of its four potential donor atoms of which two are sterically available for metal chelation (Zangrando *et al.*, 2015; Núñez-Montenegro *et al.*, 2018; Ali *et al.*, 2002). Researchers in this area have been continuing the synthesis of new nitrogen-sulphur donor ligands through Schiff base condensation with aldehyde and ketones (Tarafder *et al.*, 2001; Islam & Bashar, 2015; Kudrat-E-Zahan & Islam, 2015). The properties of these complexes can be greatly modified through the introduction of organic substituents, and a number of synthesized complexes continues to increase because of the intriguing observation that different ligands show possible different properties, although they differ only slightly in their molecular structures (Ali & Tarafdar, 1977; Ali *et al.*, 1987; Hazari *et al.*, 1996, Ali *et al.*, 1984). Transition metal complexes of N,S donor Schiff bases are widely studied because of their potential biological activities which are suitable for therapeutic use (How *et al.*, 2008; Ravooof *et al.*, 2007; Mun *et al.*, 2008; Garoufis *et al.*, 2009). Therefore, bearing in mind the importance of dithiocarbamate, we report a novel series of complexes of Ni(II), Cu(II), Zn(II) and Cd(II) with benzyl 2-(2,3-dimethoxybenzylidene)hydrazinecarbodithioate resulting from 2,3-

dimethoxybenzaldehyde and S-benzylthiocarbamate. These metal-complexes are tested for biological activity with pathogenic and non- pathogenic bacteria. The antibacterial activities of the complexes have been studied against two pathogenic bacteria (viz. *Escherichia coli*, *Shigella sonne*) and one non-pathogenic bacteria (*Bacillus subtilis*).

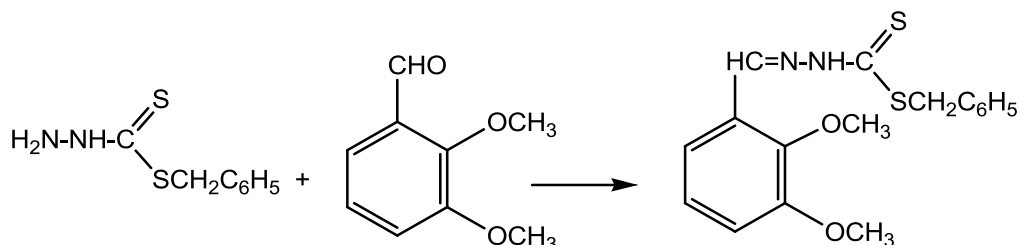
2. Experimental Data

Reagents, Chemicals, and measurements: All the reagents used were of chemical grade purity and the solvents were purified under standard procedures. Experiments and analyses were carried out at Rajshahi University, Bangladesh at the department of Chemistry. Infrared spectra were recorded using SHIMADZU FTIR-8101 infrared spectrophotometer with KBr disk, from 4000–225 cm^{-1} in the Science Lab of Rajshahi University, Rajshahi. The $^1\text{H-NMR}$ spectra were taken from BCSIR, Dhaka, Bangladesh. Conductivity measurements of the present complexes were carried out in dimethyl sulfoxide (DMSO). 10^{-3} M solutions of the complexes were employed for this purpose. The conductance measurements were made at room temperature using a type CG 857 No. 71798 SCHOTT-GERATE GmbH, Germany, digital conductivity meter and a dip type cell with a polarized electrode. The UV–vis spectrum, in DMSO were recorded on Shimadzu UV-160 spectrophotometer over the 200-900 nm range. The ESI-MS spectra were performed using a Shimadzu–Ge–Ms–Qp 100 EX mass spectrometer using the direct inlet system. Biological activities were evaluated in department of Pharmacy, University of Rajshahi. Magnetic measurements were performed using a Sherwood Scientific magnetic susceptibility balance. The metal contents were determined gravimetrically. Microanalyses (C, H, N) were executed by using a MLW-CHN micro analyser.

Synthesis: S-benzylthiocarbamate (SBDTC) was prepared according to the previous method proposed by Ali & Tarafdar et al. (1977).

Preparation of the Schiff base 2,3-dimethoxybenzaldehyde Schiff base of SBDTC (NSH):

Ethanol (50-60 ml) solution of SBDTC (1.98 g, 10 mmol) was mixed with 2, 3-dimethoxybenzaldehyde (1.66 g, 10 mmol) in hot absolute ethanol (40 ml). The mixture was refluxed for 40 min. The yellow precipitate, which formed was separated and dried *in vacuo* over anhydrous CaCl_2 m.p. 178 $^\circ\text{C}$ (Scheme1). $^1\text{H NMR}$ spectrum, δ , ppm: 2.0 s (NH), 8.74 s (CH), 7.33 d(CH), 4.42 d(CH₂), 3.83 s(CH₃). *Anal. Calc.* For $\text{C}_{17}\text{H}_{18}\text{N}_2\text{O}_2\text{S}_2$: Calculated, %: C, 58.90; H, 5.20; N, 8.09; O, 9.24; S, 18.53. Found, %: C, 58.93; H, 5.24; N, 8.09; O, 9.22; S, 18.51.

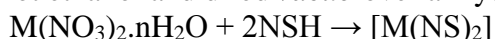


Scheme 1. Benzyl-2-(2,3-dimethoxybenzylidene)hydrazinecarbodithioate (NSH) Schiff Base derived from SBDTC

3. Results and Discussions

Synthesis of the complexes:

Benzyl-2-(2,3-dimethoxybenzylidene)hydrazinecarbodithioate (NSH) (Scheme.1) (0.321 g, 1 mmol) in hot absolute ethanol (70 ml) was added to the metal solution of the hydrated metal salt, [Ni(NO₃)₂.6H₂O] (0.145 g, 0.5 mmol); [Cu(NO₃)₂.3H₂O] (0.121g,0.5mmol); [Zn(NO₃)₂.6H₂O]. [0.148g, 0.5 mmol] and [Cd(NO₃)₂.4 H₂O] (0.154 g, 0.5 mmol) dissolved in absolute ethanol (20 ml). Each mixture was then refluxed for 50 mins and then cooled to room temperature. The precipitate was filtered off and washed with hot ethanol and dried *vacuo* over anhydrous CaCl₂.



Where, M = Ni(II), Cu(II) and Cd(II), Zn(II); NSH = 2, 3-Dimethoxybenzaldehyde Schiff base of SBDTC.

[Ni(II)(NS)₂] : Yield: 75%, Color: brown, mp: 178°C,, Anal. Calc. For C₁₇H₁₈N₂O₂S₂Ni: Calculated, %: C, 50.42; H, 4.67; N, 6.27; O, 7.54; S, 15.14; Ni, 14.84. Found, %: C, 51.03; H, 4.14; N, 6.28; O, 7.49; S, 15.66; Ni, 15.17.

[Cu(II)(NS)₂] : Yield: 72%, Color: Reddish, mp: 153°C, Anal. Calc. For C₁₇H₁₈N₂O₂S₂Cu: Calculated, %: C, 49.48; H, 4.44; N, 6.43; O, 7.48; S, 15.14; Cu, 15.42. Found, %: C, 50.45; H, 4.19; N, 6.27; O, 7.18; S, 15.54; Cu, 15.39.

[Zn(II)(NS)₂] : Yield: 76%, Color: Yellow, mp: 182°C: 185°C, Anal. Calc. For C₁₇H₁₈N₂O₂S₂Zn: Calculated, %: C, 49.51; H, 4.36; N, 6.79; O, 7.76; S, 15.53; Zn, 15.86. Found, %: C, 50.03; H, 4.34; N, 6.27; O, 7.19; S, 15.26; Zn, 15.37.

[Cd(II)(NS)₂] : Yield: 70%, Color: Reddish black, mp: 165°C,, Anal. Calc. For C₁₇H₁₈N₂O₂S₂Cd: Calculated, %: C, 44.36; H, 3.91; N, 6.09; O, 6.17; S, 13.91; Cd, 24.44. Found, %: C, 44.21; H, 3.24; N, 6.23; O, 6.15; S, 13.39; Cd, 24.25.

IR studies:

The IR spectrum of SBDTC (Fig. 1) showed strong intensity bands at 3360 and 3210 cm⁻¹, which were assigned to the asymmetric ν(N-H) and symmetric ν(N-H) modes of the NH₂ group, respectively. The ν(C=S) band for SBDTC appeared at 1060 cm⁻¹.

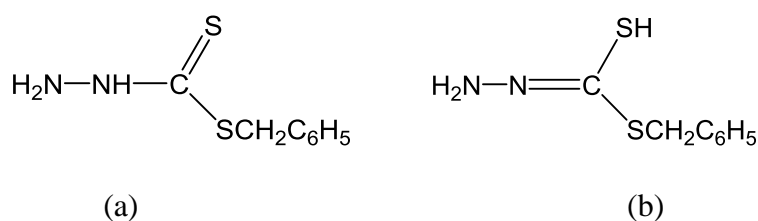


Fig. 1. (a) Thione and (b) thiol tautomeric forms of SBDTC

SBDTC has a proton adjacent to the thione group. The thione group was relatively unstable in the monomeric form and tends to turn to the more stable C-S bond by enethiolisation if there was at least one proton adjacent to the thione group. The IR spectrum of Schiff base did not display ν(S-H) stretching band at ca.2575 cm⁻¹ indicating that in the solid state it remained in the thioketo forms (1a). However, in solution, both the thion and thiol tautomeric forms (1b) are possible. In alkaline medium, the ligand deprotonates by the loss of the proton from NH group of thiol tautomeric forms (1b). Presence of the ν(C=N) group and disappearance of ν(N-H) indicates deprotonation. This

will be further evident from the absence of $\nu(\text{C}=\text{S})$ band in the complexes (Ali *et al.*, 2002; Tarafder *et al.*, 2001).

SBDTC be able to exhibit the following three modes of coordination

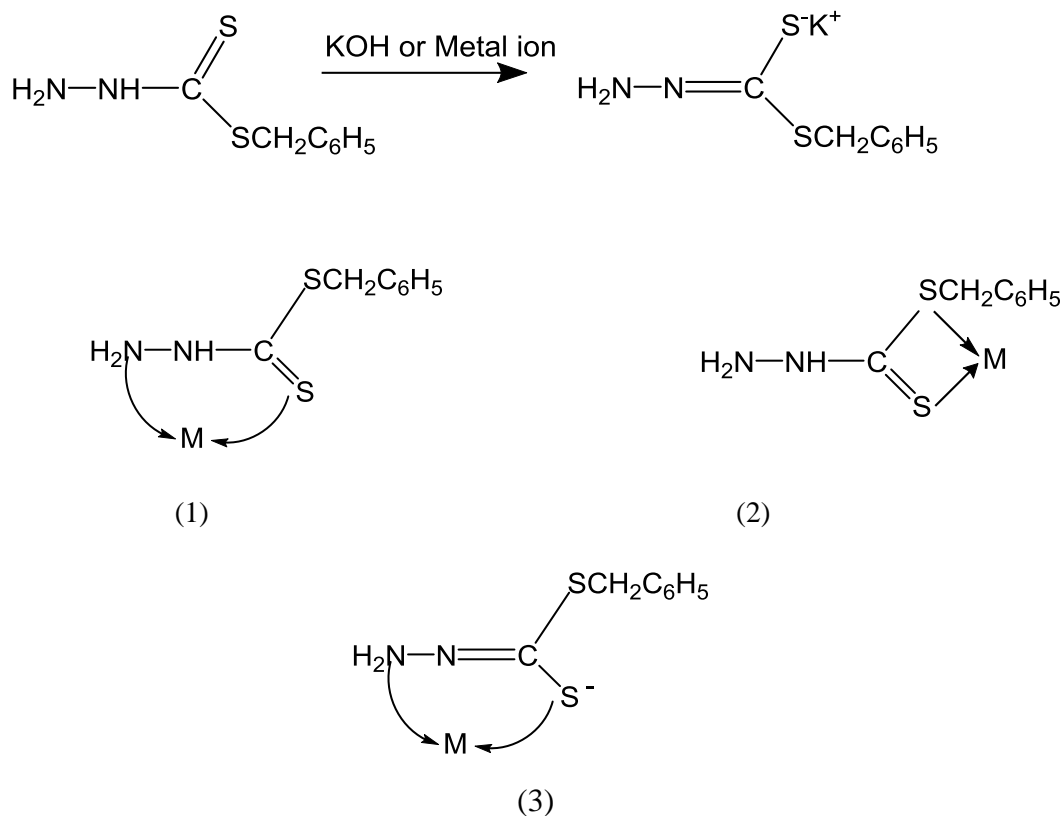


Fig. 2. Coordination modes of SBDTC

Schiff base behaves as uninegatively charged bidentated ligand by co-ordinating through the thiole sulfur and the azomethine nitrogen. The IR spectra (Table 1) of the Schiff base showed strong bands at 3210 cm^{-1} . This was attributed to the secondary amine $\nu(\text{N-H})$ mode of the free Schiff Base. The thione groups were relatively unstable in the monomeric form and tend to turn to the more stable thiole forms by enethiolization in solution (Fig.1). The absence of $\nu(\text{S-H})$ absorbance at approximately 2575 cm^{-1} indicated that in solid state, the base remained primarily in thione form. The disappearance of $\nu(\text{N-H})$ bands of the metal complexes suggested deprotonation and consequent co-ordination through the thiolate. The Schiff base also showed strong peak at 1610 cm^{-1} , which was assigned to the $\nu(\text{C}=\text{N})$. In the metal complexes, this stretching band shifted to lower frequencies, due to the lowering of the $\text{C}=\text{N}$ bond order as a result of the metal–nitrogen bond formation. The Schiff base also showed $\nu(\text{C}=\text{S})$ stretching at 1028 cm^{-1} . $\nu(\text{C}=\text{S})$ mode observed in the free ligand, disappeared in the complexes, thus supporting the above contention of thiolate bonding with metal ions. Consequently the $\nu(\text{C-S})$ mode was observed in the spectra of the complexes supporting thiolate binding of Schiff base (Kudrat-E-Zahan & Islam, 2015; Ali & Tarafdar, 1977; Bhattacharjee & Bhattacharyya, 1992).

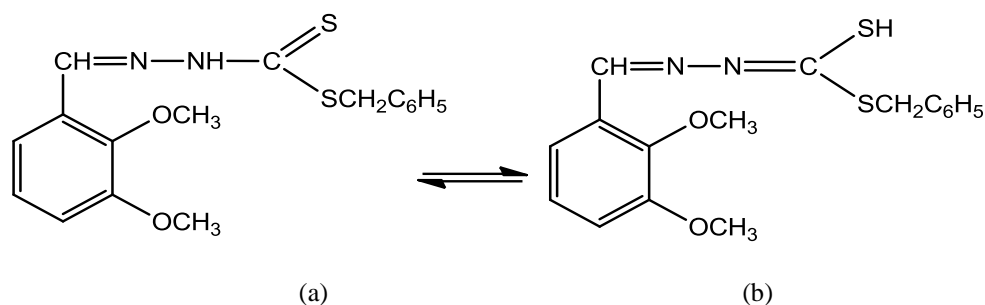


Fig. 3. (a) Thione and (b) thiol tautomeric forms of Benzyl-2-(2,3-dimethoxybenzylidene)hydrazinecarbodithioate (NSH)

The C=N stretching vibration in the free NSH Schiff base appeared at $\approx 1610\text{ cm}^{-1}$, which upon complexation, shifted towards lower frequency regions, at $\approx 1604\text{ cm}^{-1}$ (Fig.1). This lowering of C=N stretching mode on complexation supported coordination through nitrogen atom. The mode of the free Schiff base appeared at 1028 cm^{-1} . This band also disappeared in the IR spectra of the metal complexes giving evidence of coordination through the thiolate anion. The Schiff base coordinated to the metal through the thiolate sulphur and the β -nitrogen as evident from the IR spectrum showing bands at $\approx 391\text{ cm}^{-1}$ $\nu(\text{M-S})$ and $\approx 449\text{ cm}^{-1}$ $\nu(\text{M-N})$, respectively.

From the above evidence (Table.1) IR spectra for the complexes $[\text{Ni}(\text{II})(\text{NS})_2]$ $[\text{Zn}(\text{II})(\text{NS})_2]$ $[\text{Co}(\text{II})(\text{NS})_2]$ and $[\text{Cd}(\text{II})(\text{NS})_2]$ gave the characteristics bands for (M-N) and (M-S) which indicate the direct bonding between M and N and coordination bonding between M and S (Bitu *et al.*, 2020; Latif *et al.*, 2019).

Table 1. Infrared spectral bands (cm^{-1}) of the Schiff Base and metal complexes

Compounds	$\nu(\text{NH}_2)$	$\nu(\text{N-H})$	$\nu(\text{C=S})$	$\nu(\text{C=N})$	$\nu(\text{C-S})$	$\nu(\text{M-S})$	$\nu(\text{M-N})$
SBDTC	3360, 3210	3210	1060	-	-	-	-
NSH	-	3100	1091	1610	-	-	-
$[\text{Cu}(\text{II})(\text{NS})_2]$	-	-	-	1604	715	391	449
$[\text{Ni}(\text{II})(\text{NS})_2]$	-	-	-	1605	707	406	479
$[\text{Zn}(\text{II})(\text{NS})_2]$	-	-	-	1603	678	387	489
$[\text{Cd}(\text{II})(\text{NS})_2]$	-	-	-	1596	716	377	477

Table 2. Molar conductance and magnetic moment data

NSH/Complexes	Molar conductance ($\text{ohm}^{-1}\text{ cm}^2\text{ mol}^{-1}$)	μ_{eff} (B.M.)
NSH	2.30	-
$[\text{Ni}(\text{II})(\text{NS})_2]$	2.10	diamagnetic
$[\text{Cu}(\text{II})(\text{NS})_2]$	1.98	1.98
$[\text{Zn}(\text{II})(\text{NS})_2]$	3.95	3.94
$[\text{Cd}(\text{II})(\text{NS})_2]$	2.45	3.97

ESI-MS Studies:

ES-MS has been increasingly employed as a dominant structural characterization method in coordination chemistry. The spectra of the ligand and Cu(II), Ni(II), Zn(II) and Cd(II) complexes show molecular ion peaks at $m/z = 346.04$, 754.57 , 749.28 , 756.16 , and 803.56 amu respectively, which are equivalent to their molecular mass. The molecular ion peaks are in good agreement with the proposed molecular formulae indicated from microanalysis.

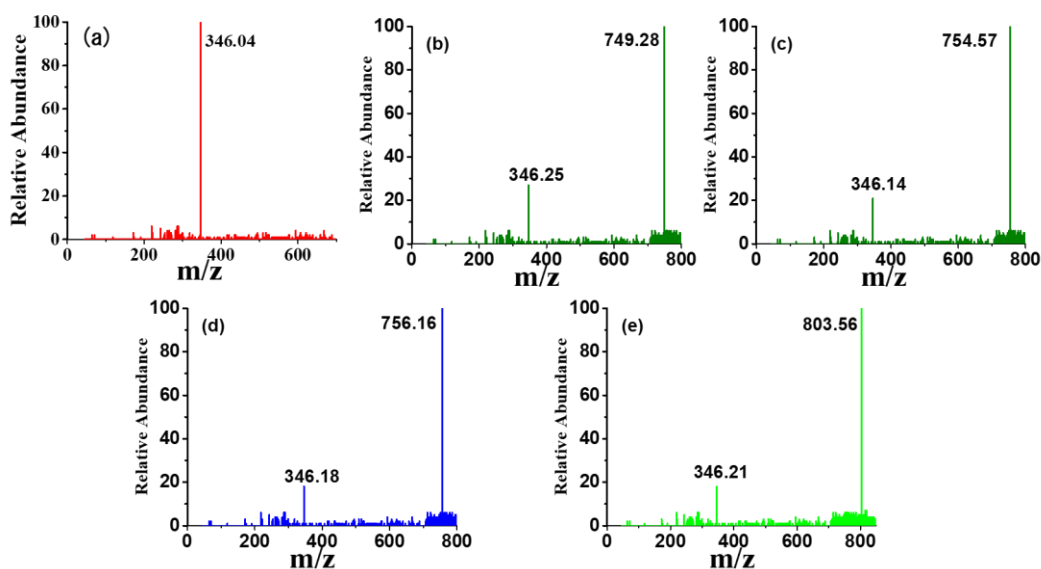


Fig. 4. ESI-MS spectra of (a) Schiff Base; (b) Cu; (c) Ni; (d) Zn and (e) Cd complex

Magnetic moment and electronic spectra:

The magnetic susceptibility measurement (Table 2) showed that complex $[\text{Ni(II)(NS)}_2]$ was diamagnetic. The diamagnetic nature of $[\text{Ni(II)(NS)}_2]$ indicated a singlet ground state characteristic of square planar Ni(II). Three bands corresponding to the transitions ${}^1A_{1g} \rightarrow {}^1A_{2g}$ (251 nm), ${}^1A_{1g} \rightarrow {}^1B_{1g}$ (340 nm) and ${}^1A_{1g} \rightarrow {}^1A_{2g}$ (420 nm) were observed in the electronic spectrum of such square-planar nickel(II) complex.

The magnetic susceptibility measurement (Table 2) showed that complex $[\text{Cu(II)(NS)}_2]$ was paramagnetic. The greenish brown paramagnetic copper complex gave a magnetic moment of 1.98 B.M. corresponding to one unpaired electron. The UV-vis spectrum of the complex showed d-d bands at 250 nm, 294 nm and 373 nm arising from the ${}^2B_{1g} \rightarrow {}^2A_{1g}$ and ${}^2B_{1g} \rightarrow {}^2E_{1g}$ transitions, respectively, characteristic of a square-planar stereochemistry. The band at 373 nm was presumably caused by charge transfer (Gray & Ballhausen, 1963; Jørgensen, 1962; Figgis, 1967).

The complex $[\text{Zn(II)(NS)}_2]$ has a room-temperature magnetic moment of about 3.94 B.M., indicative of tetrahedral geometry. The electronic spectra of $[\text{Zn(II)(NS)}_2]$ showed three bands at 242, 296 and 341 nm (Islam & Bashar, 2015; Bitu *et al.*, 2020; Latif *et al.*, 2019).

The effective magnetic moment of cadmium complex at room-temperature is shown in Table 2. The electronic spectra of $[\text{Cd(II)(NS)}_2]$ complex contributed two intense bands at 359 and 246 nm corresponding to the transition, ${}^4A_{2g}(F) \rightarrow {}^4T_{1g}(P)$ and charge transfer band, respectively (Malik *et al.*, 1973; Duan *et al.*, 1997; Singh *et al.*, 2009). This spectral data indicated tetrahedral stereochemistry of Cd(II) complex. The above observations indicated $[\text{Ni(II)(NS)}_2]$ and $[\text{Cu(II)(NS)}_2]$ have square planar

structure with two ligands satisfying the four stereochemical sites (Fig.5), while $[\text{Zn(II)(NS)}_2]$ and $[\text{Cd(II)(NS)}_2]$ are tetrahedral in geometry (Fig. 6).

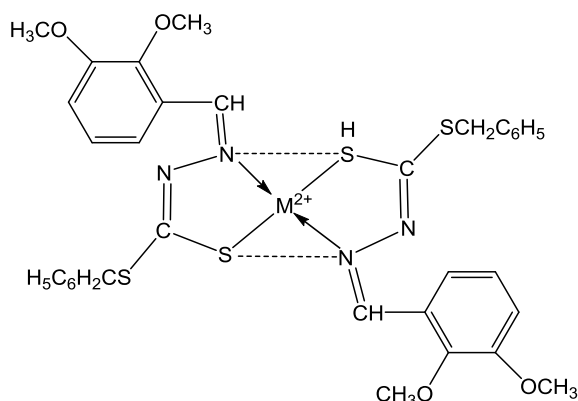


Fig. 5. Square planar structure of Ni(II) and Cu(II) complexes

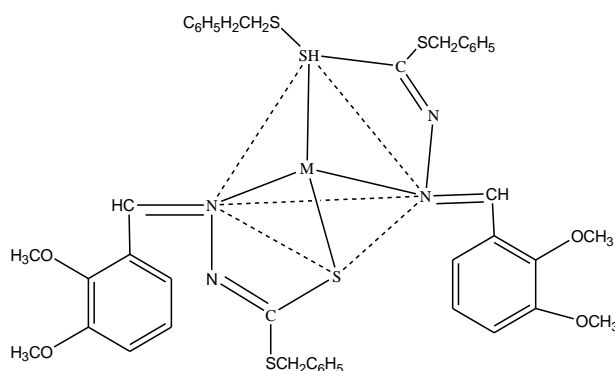


Fig. 6. Tetrahedral structure of Zn(II) and Cd(II) complexes

Antibacterial activity: The synthesized compounds were tested for antibacterial activity against three pathogenic bacteria, two gram-negative (*E. coli* and *S. sonnei*) and one gram-positive (*B. subtilis*). Disc diffusion method is used in this study which is a widely accepted procedure for in vitro investigation of the susceptibility of microorganisms to the compounds, so this method was adopted (Latif, *et al.*, 2019; Hossain *et al.*, 1996; Manan *et al.*, 2012). The metal complexes yielded clear inhibition zones around the discs with all the three test bacteria (Table 3, Fig.7.). The complexes demonstrated moderate to strong activities against both gram-positive and gram-negative bacteria compared to the antibacterial standard Kanamycin (Table 3).

Table 3. Test results of antibacterial activity

Compound	Diameter of the inhibition zone, mm		
	<i>E. coli</i>	<i>S. sonnei</i>	<i>B. subtilis</i>
NSH	12	14	13
$[\text{Ni(II)(NS)}_2]$	16	15	17
$[\text{Cu(II)(NS)}_2]$	17	16	15
$[\text{Zn(II)(NS)}_2]$	11	13	14
$[\text{Cd(II)(NS)}_2]$	13	12	16
Kanamycin	20	20	22

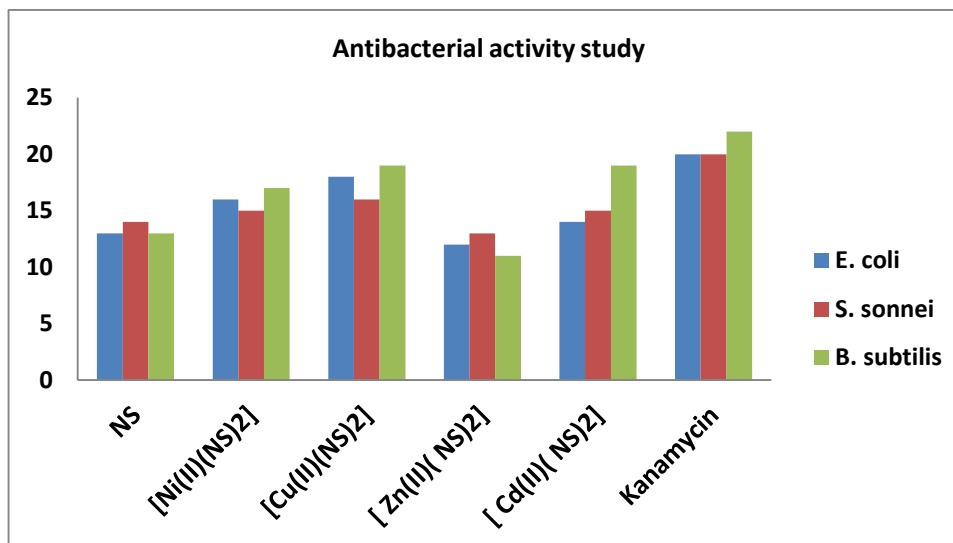


Fig. 7. Pictorial presentation of antibacterial activity assays

4. Conclusions

Schiff base benzyl 2-(2,3-dimethoxybenzylidene)hydrazinecarbodithioate, derived from the condensation of 2,3-dimethoxybenzaldehyde with S-benzylthiocarbamate acts as a uninegative bidentate ligand with its NS donor set to form neutral bischelated Ni(II), Cu(II), Zn(II) and Cd(II) complexes. The Ni(II) and Cu(II) complexes have a square-planar geometry, while the Zn(II) and Cd(II) complexes are tetrahedral. The ligand and its metal complexes showed significant antimicrobial activity against two gram-negative (*E. coli* and *S. sonnei*) and one gram-positive (*B. subtilis*) bacteria compared to standard Kanamycin.

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