



# Thermal and Spectral Characterization of Cr(III), Co(II) and Cd(II) Metal Complexes Containing Bis-Imine Novel Schiff Base Ligand Towards Potential Biological Application

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**Abstract:** Metal complexes of Cr(III), Co(II) and Cd(II) ions were synthesized with a ONS containing Schiff base ligand, 2-bis(2-oxoindolin-3-ylidene)hydrazinecarbothioamide which was derived from the condensation reaction of thiosemicarbazide and isatin. The ligand and complexes were isolated from the reaction in the solid form and characterized by IR, UV-Visible, Thermal analysis and some physical measurements. Spectroscopic evidence indicated that the Schiff base behaved as ONS coordinating hexadentate chelating agent. Magnetic susceptibility data coupled with electronic spectra suggested a distorted octahedral structure of the complexes. The overall reaction was monitored by TLC and UV-Visible spectral analysis. The Schiff base and their metal complexes have been shown moderate to strong microbial activity.

**Keywords:** Transition Metal Complex, IR, UV-Visible Spectra Analysis, Antibacterial Activity, Schiff Base, TGA and DTG

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## 1. Introduction

Schiff bases are condensation products of primary amines and carbonyl compounds. They were first discovered by a German chemist, Nobel Prize winner, Hugo Schiff. Structurally, Schiff base (also known as imine or azomethine) is an analogue of a ketone or aldehyde in which the carbonyl group (C=O) has been replaced by an imine or azomethine group. The versatility and flexibility of Isatin based Schiff base compounds having, acyl, aroyl and heteroacroyl Schiff bases have additional donor sites >C=O, >C=N-, etc. It has made the Schiff base to act as good complexing agents that form a variety of complexes with various transition and inner transition metals and has emphasized the attention of many researchers [1]. Bis-Imines and their derivatives have outstanding characteristics that they can increase molecular

conjugate system and enhance photoelectric property. Bis-Schiff base ligands and their coordination compounds having multifunctional groups play an important role in the areas of stereochemistry, structure of science, spectroscopy, magnetic fields [2]. In recent years, sulfur containing ligands such as dithiocarbamates and thiosemicarbazones and their transition metal complexes have received more attention in the area of medicinal chemistry, due to their pharmacological properties, such as antiviral, antibacterial, antifungal, antiparasitic, and antitumor activities [3]. In addition, due to their bonding modes, biological implications, structural diversity, and ion-sensing ability have received thiosemicarbazones as great interest in chemistry [4]. Moreover, Schiff bases are regarded as privileged ligands [5]. Schiff base complexes with different transition metals can act as catalysts for various reactions [6, 7]. The metal complexes of thiosemicarbazone dramatically increase the biological activities such as

antibacterial, antifungal, anti HIV, and anti-inflammatory [8]. Thiosemicarbazones and their metal complexes are also applicable in the field of material sciences such as nonlinear optical (NLO) [9], electrochemical sensing [10], and Langmuir film [11]. Schiff bases derived from isatin exhibit many neurophysiological and neuropharmacological effects like antimicrobial, antiviral, anticonvulsant, anticancer, antimycobacterial, antimalarial, cysticidal, herbicidal and anti-inflammatory activity [12–14]. They also exhibit anti-HIV, antiprotozoal and antihelminthic activities [15–17]. Recently, Cd(II), Ni(II), Co(II) and Zr(IV) metal complexes of bis-imine Schiff base ligand derived from diethylenetriamine and isatin was reported from our laboratory [18]. In view of the interesting application of these type of Schiff base ligands and their metal complexes inspired us for the synthesis, characterization and biological activity studies of metal complexes with Schiff base ligands derived from isatin and thiosemicarbazide.

## 2. Experimental

### 2.1. Reagents and Chemicals

All the reagents used were of analar grade or chemically pure grade. All metal(II) salts were used as chloride and nitrate. The solvents such as ethanol, methanol, chloroform, diethyl ether, petroleum ether, DMSO (dimethyl sulfoxide), dichloromethane and acetonitrile were purified by known procedure [19].

### 2.2. Physical Measurement

The melting point or the decomposition temperature of all the prepared ligand and metal complexes were observed in an electro thermal melting point apparatus model No. AZ6512.

Vibrational spectra (IR) were recorded with SHIMADZU-8400, FTIR spectrophotometer, (Japan), in the range 4000-400  $\text{cm}^{-1}$  with a KBr disc as reference. UV-Visible spectra of the complexes in DMF (0.0005 molar) were recorded in the region 200-800 nm on a THERMOELECTRON NICOLET evolution 300 UV-Visible spectrophotometer. The SHERWOOD SCIENTIFIC Magnetic Susceptibility Balance that following the Gouy method were used to measure the magnetic moment of the solid complexes. The electrical conductance measurements were made at room temperature in freshly prepared solution ( $10^{-3}$  M) in DMF using a WPACM35 conductivity meter and a dip-cell with a platinum electrode. The thermogravimetric analysis (TGA) was performed on Perkin Elmer Simultaneous Thermal Analyzer, STA-8000. The purity of the ligand and metal complexes were tested by Thin Layer Chromatography (TLC).

### 2.3. Preparation of Schiff Base [SB]

Isatin/indoline-2,3-dione (2.94 g, 20 mmol) dissolved in absolute ethanol (30 mL) was added slowly to a constant stirring ethanolic solution of thiosemicarbazide (0.91 g, 10 mmol) containing 5 ml of conc.  $\text{H}_2\text{SO}_4$ . The reaction mixture was refluxed for 6h. On cooling, a solid orange product was formed which was filtered, washed with ethanol and diethyl ether and dried in vacuum over anhydrous  $\text{CaCl}_2$ . The synthesized reaction of ligand and complexes were monitored by TLC using petroleum ether, ethyl acetate, toluene and methanol as solvent. The solid product obtained was found to be soluble in methanol, DMF and DMSO and insoluble in ethanol, acetone, diethyl ether, petroleum ether and isopropanol. The structure of Schiff base was shown in fig-1.

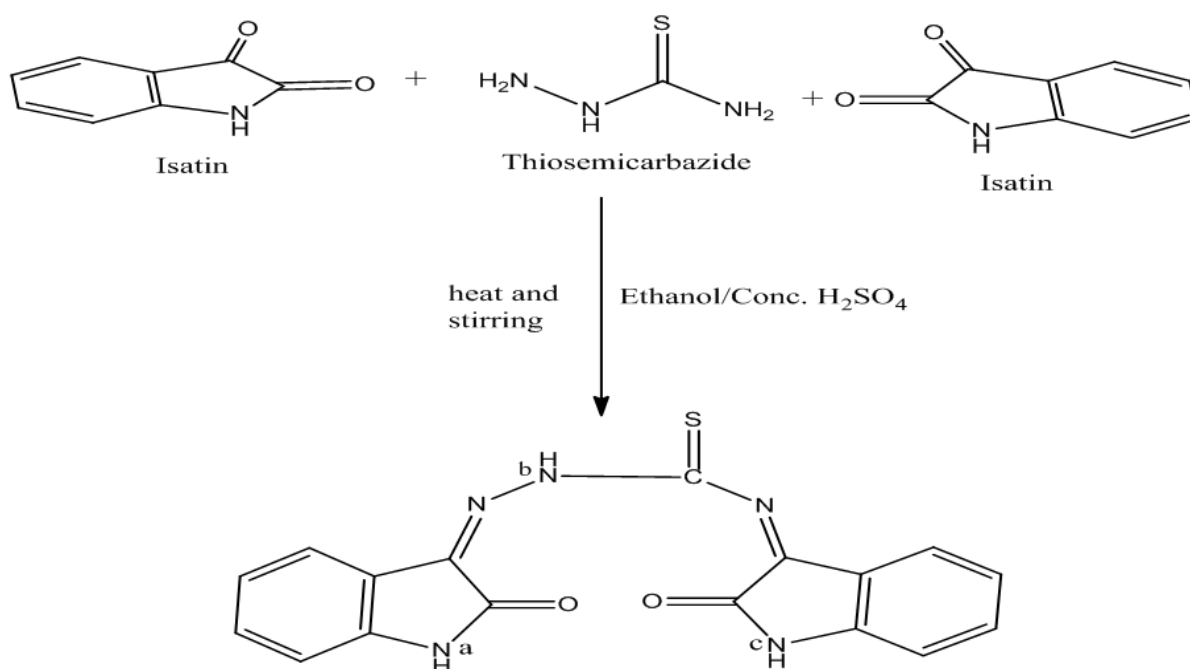


Fig. 1. Structure of Schiff base [SB] where, [SB] =  $[\text{C}_{17}\text{H}_{11}\text{N}_5\text{O}_2\text{S}]$ .

## 2.4. Preparation Procedure of Schiff Base [SB] Metal Complexes

The complexes have the general formula  $[M(SB)]$ ; where  $M = \text{Cr(III)}$ ,  $\text{Co(II)}$ , and  $\text{Cd(II)}$  ions and SB = Synthesized Schiff base ligand (Fig-1). Ethanolic solution (20 mL) of Chromium(III) nitrate nona hydrate (0.400g, 1 mmol)/Cobalt(II) chloride hexahydrate (0.2378 g, 1 mmol)/Cadmium(II) chloride mono hydrate (0.201 g, 1 mmol) was taken in a two necked round bottom flask and kept on magnetic stirring. A warm ethanolic solution (20 mL) of

prepared Schiff base ligand (0.349 g, 1mmole) was added drop wise and stirred with heating for 4h. On cooling, precipitates were formed which were filtered, washed with ethanol, acetone, and diethyl ether and dried in vacuum desiccators over anhydrous  $\text{CaCl}_2$ . The purity of each complexes were tested by TLC using petroleum ether, ethyl acetate, toluene and methanol as solvents. The complexes were soluble in DMSO and DMF. The structure of complex was shown in fig-2.

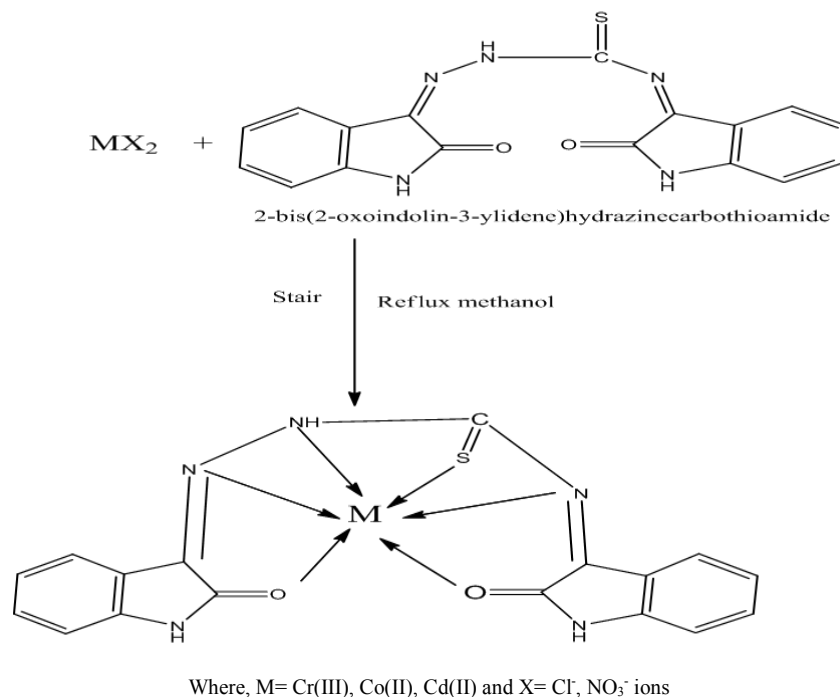


Fig. 2. Proposed Structure of Schiff base metal complexes.

## 3. Result and Discussion

### 3.1. Physical Properties

Some physical properties of the Schiff base ligand and its metal complexes such as melting point, color, magnetic moment etc are shown in table-1. The complexes are intensely colored, powdered solids, which decomposes above  $300^\circ\text{C}$ . Molar conductance values in DMSO ( $10^{-3}\text{M}$ ) showed low values (3-5  $\mu\text{S/cm}$ ) indicating [20] them to be non-electrolyte.

Table 1. Physical and analytical data of the Schiff base and metal complexes.

Compound/Mol. formula	Color yield (%)	Melting point / $^\circ\text{C}$	Conductivity /( $\mu\text{S/cm}$ )
[SB]			
$[\text{C}_{17}\text{H}_{11}\text{N}_5\text{O}_2\text{S}]$	Orange	73	140-145
$[\text{C}_{17}\text{H}_{11}\text{CrN}_5\text{O}_2\text{S}]$	Greenish	64	>300
$[\text{C}_{17}\text{H}_{11}\text{CoN}_5\text{O}_2\text{S}]$	Coffee	62	>300
$[\text{C}_{17}\text{H}_{11}\text{CdN}_5\text{O}_2\text{S}]$	Orange	71	>300

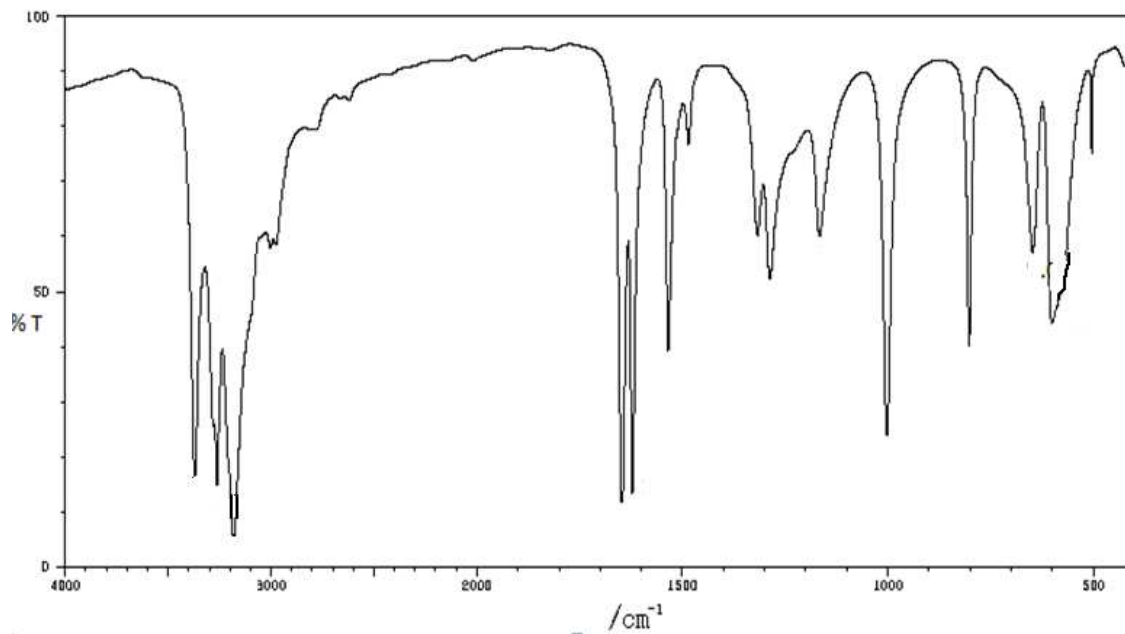
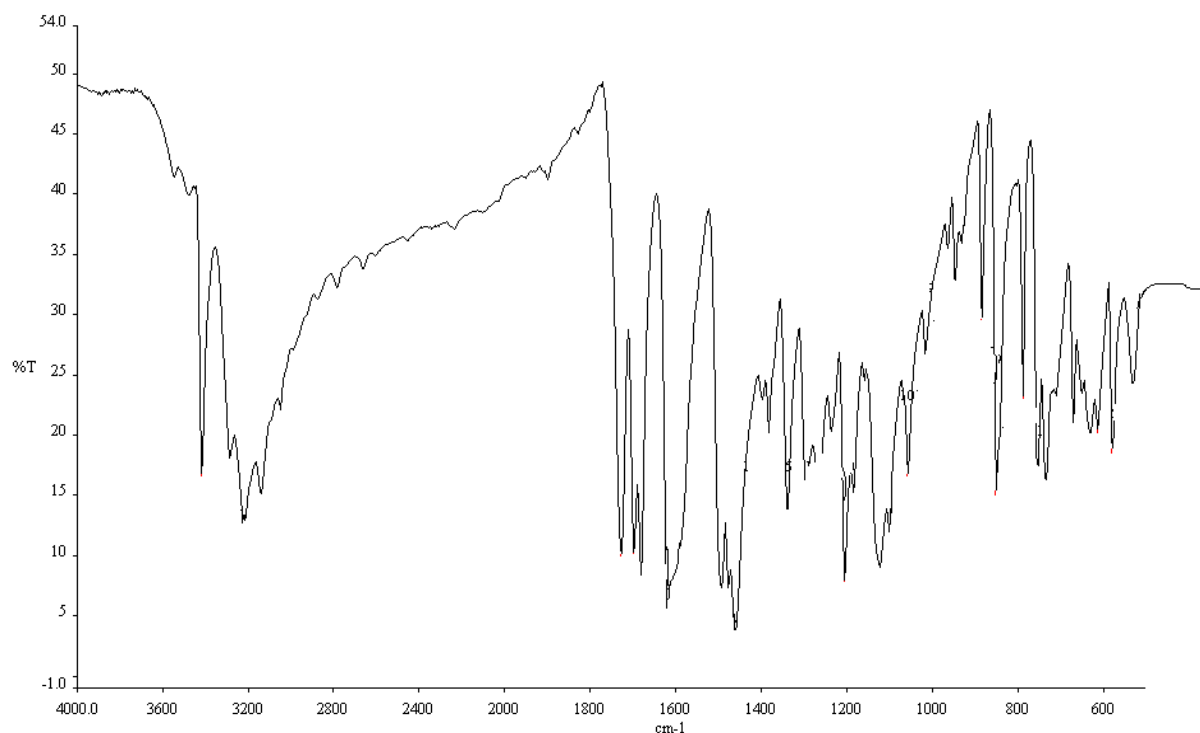
### 3.2. Infrared Spectral Analysis

The observed IR-absorption bands  $3419, 3222$  and  $3142\text{ cm}^{-1}$  due to  $^a\text{N-H}$ ,  $^b\text{N-H}$  and  $^c\text{N-H}$  respectively in the free Schiff base ligand as shown (fig-1). The strong band  $1610\text{ cm}^{-1}$  was assigned to  $\nu(\text{C=N})$  in the Schiff base ligand represented that free  $-\text{NH}_2$  group of thiosemicarbazide was converted to imine group. The band  $1727\text{ cm}^{-1}$  was assigned to  $\nu(\text{C=O})$  in Schiff base confirmed that carbonyl group of isatin was present in ligand. The strong band  $1124\text{ cm}^{-1}$  for  $\nu(\text{C=S})$  indicated C=S bond was present in the Schiff base ligand [21]. During complexation the band  $3222\text{ cm}^{-1}$  for  $^b\text{N-H}$  shifted to lower absorption frequency evident that it was coordinated to metal atoms. The shifting of bands from  $1727, 1610$  and  $1124\text{ cm}^{-1}$  to lower absorption frequency suggested that C=O, C=N and C=S groups respectively was coordinated to central metal atoms. Also the appearance of new low absorption bands at (482-493), (532-604), (404-434) assigned to  $\nu(\text{M-N})$ ,  $\nu(\text{M-O})$  and  $\nu(\text{M-S})$  respectively [22] confirmed that O, N and S atoms were coordinated to central metal atoms.

**Table 2.** Selected IR spectral data of the Schiff base and metal complexes.

Ligand/complex	IR( $\text{cm}^{-1}$ )						
	$\nu(\text{N-H})$	$\nu(\text{C=N})$	$\nu(\text{C=O})$	$\nu(\text{C=S})$	$\nu(\text{M-N})$	$\nu(\text{M-O})$	$\nu(\text{M-S})$
$[\text{C}_{17}\text{H}_{11}\text{N}_5\text{O}_2\text{S}]$	3222	1610	1727	1124	-	-	-
$[\text{C}_{17}\text{H}_{11}\text{CrN}_5\text{O}_2\text{S}]$	3216	1606	1680	1116	487	532	434
$[\text{C}_{17}\text{H}_{11}\text{CoN}_5\text{O}_2\text{S}]$	3214	1603	1685	1108	482	581	428
$[\text{C}_{17}\text{H}_{11}\text{CdN}_5\text{O}_2\text{S}]$	3207	1570	1676	1106	493	604	404

Where, [SB]= Schiff base ligand  $[\text{C}_{17}\text{H}_{11}\text{N}_5\text{O}_2\text{S}]$

**Fig. 3.** IR Spectra of thiosemicarbazide.**Fig. 4.** IR Spectra of Schiff base ligand  $[\text{C}_{17}\text{H}_{11}\text{N}_5\text{O}_2\text{S}]$ .

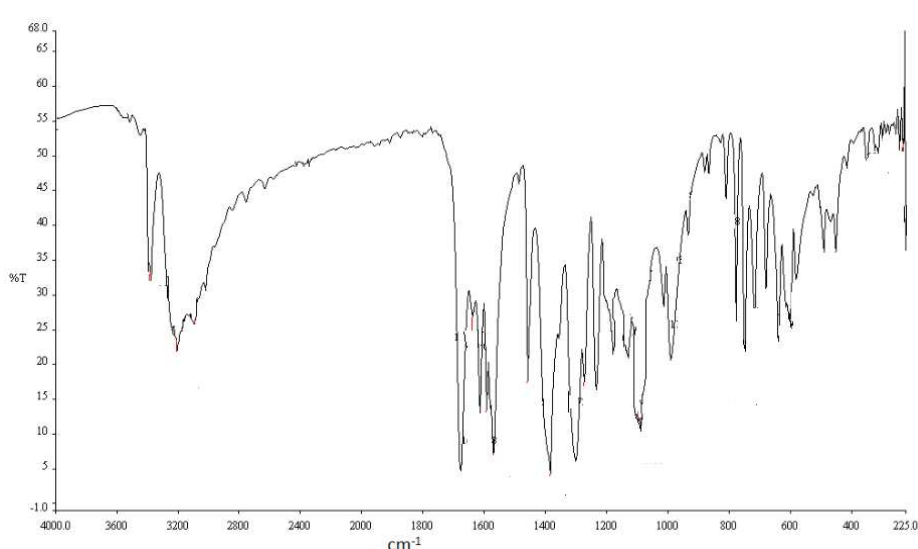


Fig. 5. IR Spectra of  $[C_{17}H_{11}CdN_3O_2S]$  complex.

### 3.3. Magnetic Moment and UV-Visible Spectral Analysis

The spectral data of both Schiff base ligand 2-bis(2-oxindolin-3-ylidene) hydrazine carbothioamide and their metal complexes were taken in DMF. The magnetic moment and UV-Visible spectral data was recorded in Table-3. At room temperature magnetic moment value of Cr(III) complex was found to 1.53 B. M representing one unpaired electron per Cr(III) ion. Electronic spectra of Cr(III) in DMF showed bands at 13,321–19,582 and 25,649–27,678  $cm^{-1}$  which may be assigned to  $4A_{2g} \longrightarrow 4T_{2g}(F)$  and  $4A_{2g} \longrightarrow 4T_{2g}(F)$  respectively [21]. similarly, Co(II) complexes was found to

4.41 B. M indicative of three unpaired electron per Co(II) ion attaining an octahedral environment, On the other hand Cd(II) complex showed zero magnetic moment that correspond to no unpaired electron per metal ion and showed absorption bands 430 nm that suggested octahedral geometry of the complexes. The electronic spectra of the Co(II) complex showed three bands observed at 15329 - 15496  $cm^{-1}$ , 24318 - 24547  $cm^{-1}$  and 2662 - 26853  $cm^{-1}$  which may be assigned to  $4A_{2g} \longrightarrow 4T_{1g}(F)$ ,  $4A_{2g} \longrightarrow 4T_{2g}(F)$  and  $4A_{2g} \longrightarrow 4T_{1g}(P)$  respectively [23].

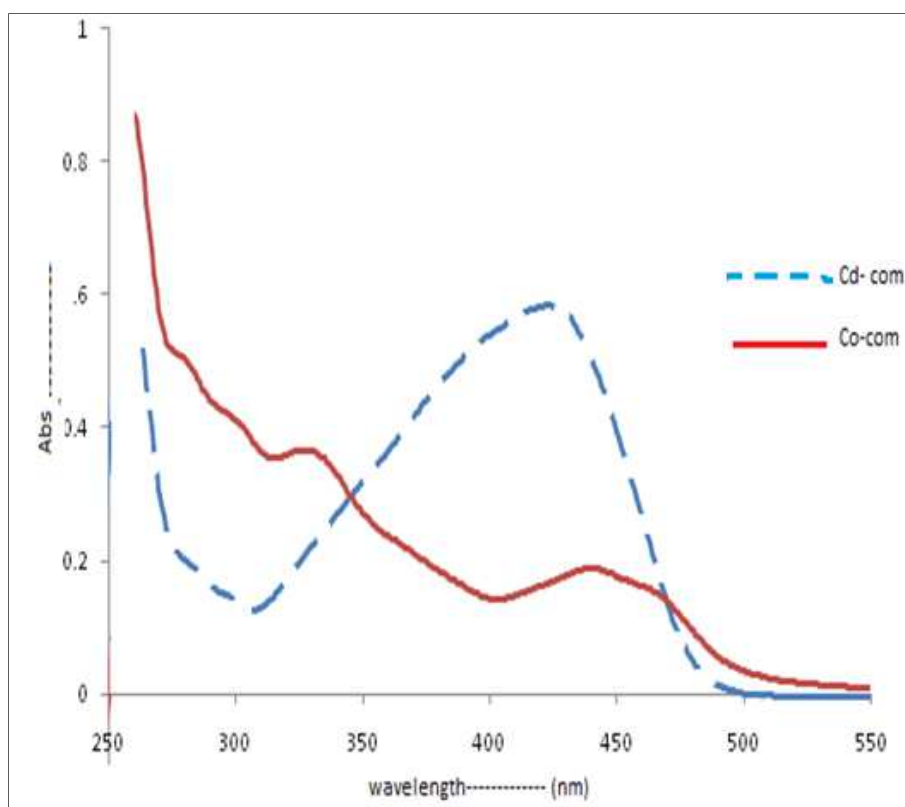


Fig. 6. UV-Visible Spectra of Cd(II) and Co(II) complexes.

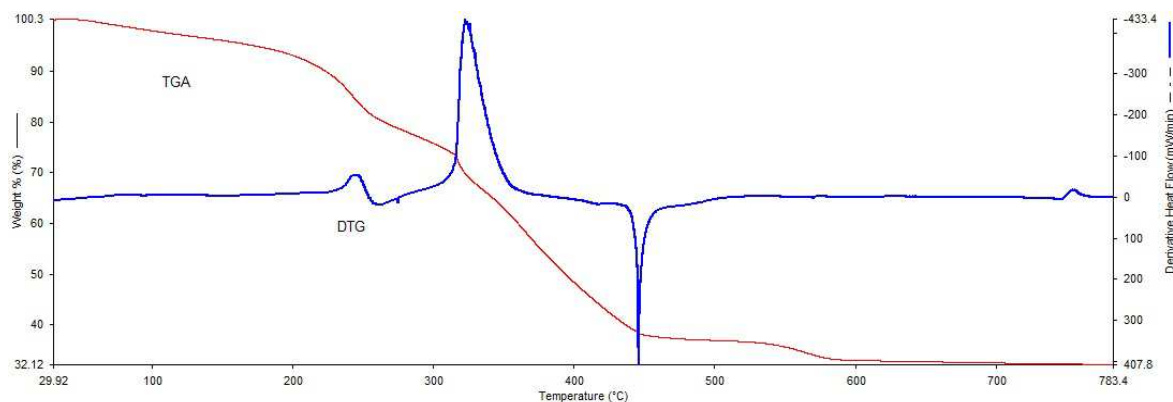
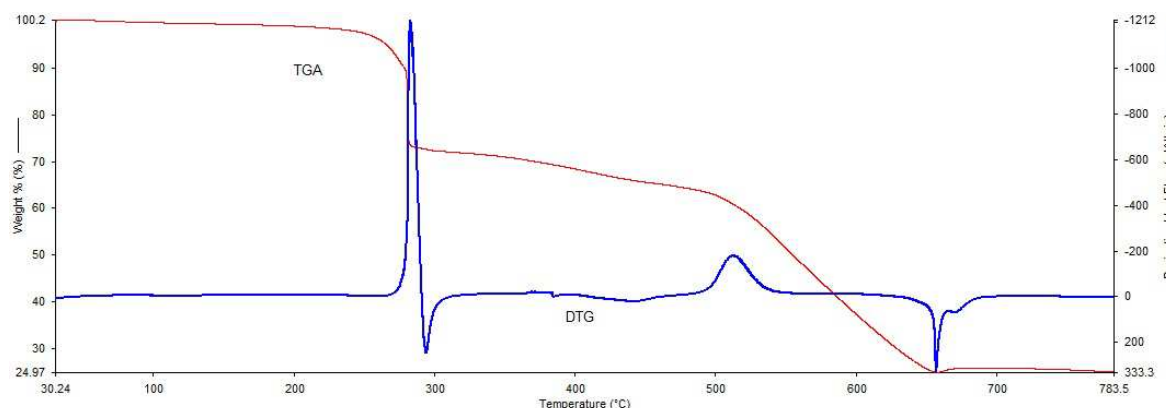
**Table 3.** Magnetic moment and UV-Visible spectral components of Metal complexes.

Complexes	$\mu_{\text{eff}}$ (B.M)	$\lambda_{\text{max}}$ ( $\text{cm}^{-1}$ )	Assignment
[C <sub>17</sub> H <sub>11</sub> CrN <sub>5</sub> O <sub>2</sub> S]	1.53	13,321–19,582 25,649–27,678	6A <sub>1g</sub> → 4T <sub>1g</sub> 6A <sub>1g</sub> → 4T <sub>2g</sub>
[C <sub>17</sub> H <sub>11</sub> CoN <sub>5</sub> O <sub>2</sub> S]	4.3	15329 – 15496 24318 – 24547	4A <sub>2</sub> → 4T <sub>1g</sub> (F) 4A <sub>2g</sub> → 4T <sub>2g</sub> (F)
[C <sub>17</sub> H <sub>11</sub> CdN <sub>5</sub> O <sub>2</sub> S]	Diamagnetic	2662 – 26853 420(nm)	4A <sub>2g</sub> → 4T <sub>1g</sub> (P) CT

### 3.4. Thermogravimetric Analysis

TGA was carried out for solid Cr(III), Co(II) and Cd(II) metal complexes under N<sub>2</sub> flow. The heating rate was suitably controlled at 30°C min<sup>-1</sup> and the weight loss was measured from the ambient temperature up to 800°C. The TGA and DTG curve of Cr(III) complex was shown in fig-7, the curve indicated that the complex was decomposed into 3 or 4 main steps. Where, the 1<sup>st</sup> step involves the removal of one molecule of water (calculated 4.29%, experimental 4.12% weight) at temperature range 52-180°C [23, 24]. The part of ligand –NH-CS– was decomposed between temperature range 190-263°C (calculated 10.26%, experimental 10.01% weight). The major fragmentation was occurred at temperature range 273-454°C that suggested the decomposition of ligand part –C<sub>8</sub>H<sub>5</sub>N<sub>2</sub>O from the complex (calculated 72.31%, experimental 65.49% weight) at 3<sup>rd</sup> steps of decomposition [25-26]. In the last step at above 630°C temperature the complex was completely decomposed and removed as Cr/CrO (calculated 12.96%, experimental

10.34% weight). From the TGA and DTG curve of [C<sub>17</sub>H<sub>11</sub> CoN<sub>5</sub>O<sub>2</sub>S] complex (fig-8) indicated that the complex was stable up to 280°C and decomposition was taken place into three main steps. In 1<sup>st</sup> step of decomposition the part of ligand –C<sub>3</sub>H<sub>3</sub>N<sub>3</sub>S– (calculated 27.27%, experimental 26.39% weight) between 278-297°C were decomposed. At temperature range 405-650°C the rest part of ligand –C<sub>7</sub>H<sub>5</sub>NO– (calculated 58.47%, Experimental 51.95% weight) was removed. At above 700°C temperature the complex was decomposed completely and removed as Co/CoO [23]. The [C<sub>17</sub>H<sub>11</sub> CdN<sub>5</sub>O<sub>2</sub>S] complex was stable up to 290°C temperature as shown in (fig-9). The TGA and DTG curve of [C<sub>17</sub>H<sub>11</sub> CdN<sub>5</sub>O<sub>2</sub>S] complex shown that the complex was decomposed into three main steps, where the major fragmentation was taken place between temperature range 300-700°C which involved the decomposition of ligand. At above 765°C temperature the complex was completely decomposed and removed as Cd/CdO. All the possible degradation pathways is shown in Fig-10.

**Fig. 7.** TGA and DTG curve of [C<sub>17</sub>H<sub>11</sub> CrN<sub>5</sub>O<sub>2</sub>S].**Fig. 8.** TGA and DTG curve of [C<sub>17</sub>H<sub>11</sub> CoN<sub>5</sub>O<sub>2</sub>S].

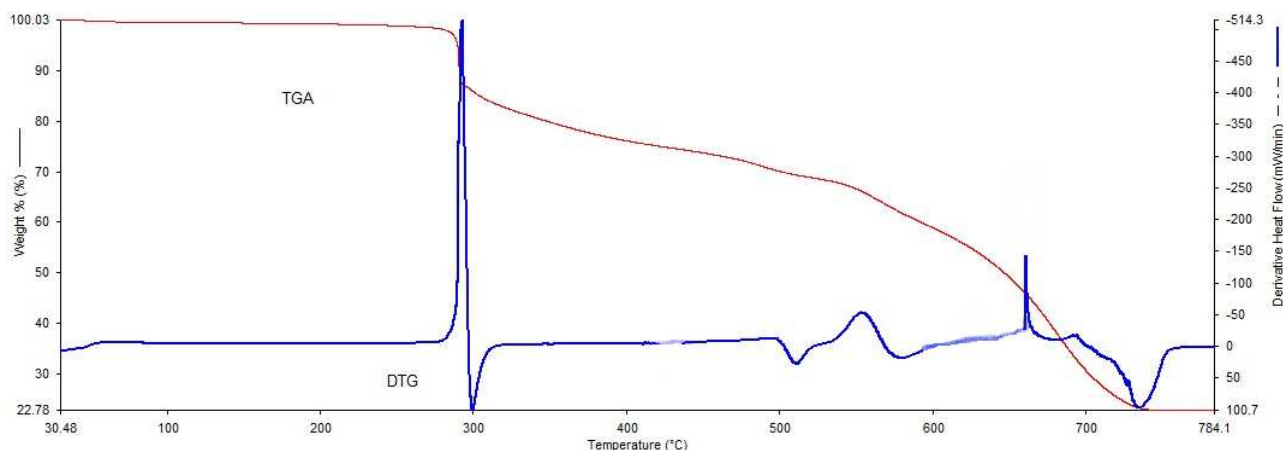


Fig. 9. TGA and DTG curve of  $[C_{17}H_{11}CdN_5O_2S]$ .

Table 4. Thermal data of Cr(III), Co(II) and Cd(II) complexes.

Complexes	Steps	Temperature Range/ °C	DTG Peak/ °C	TG mass loss% calc./found	Assignments
$[C_{17}H_{11}CrN_5O_2S].H_2O$	1 <sup>st</sup>	52-180		4.29/4.12	H <sub>2</sub> O
	2 <sup>nd</sup>	190-263	232	10.26/10.03	-NH-CS-
	3 <sup>rd</sup>	273-454		72.31/65.49	-2C <sub>8</sub> H <sub>5</sub> N <sub>2</sub> O
		>630	335	12.96/10.34	Cr/CrO
$[C_{17}H_{11}CoN_5O_2S]$	1 <sup>st</sup>	278-297	289	27.27/26.39	-C <sub>3</sub> HN <sub>3</sub> S-
	2 <sup>nd</sup>	405-650		58.47/51.95	2 C <sub>7</sub> H <sub>5</sub> NO
	3 <sup>rd</sup>	>700	526	14.21/10.38	Co/CoO
$[C_{17}H_{11}CdN_5O_2S]$	1 <sup>st</sup>	285-295		27.27/26.39	-C <sub>3</sub> HN <sub>3</sub> S-
	2 <sup>nd</sup>	500-737	287	58.47/51.95	2 C <sub>7</sub> H <sub>5</sub> NO
	3 <sup>rd</sup>	>772		24.29/20.23	Cd/CdO

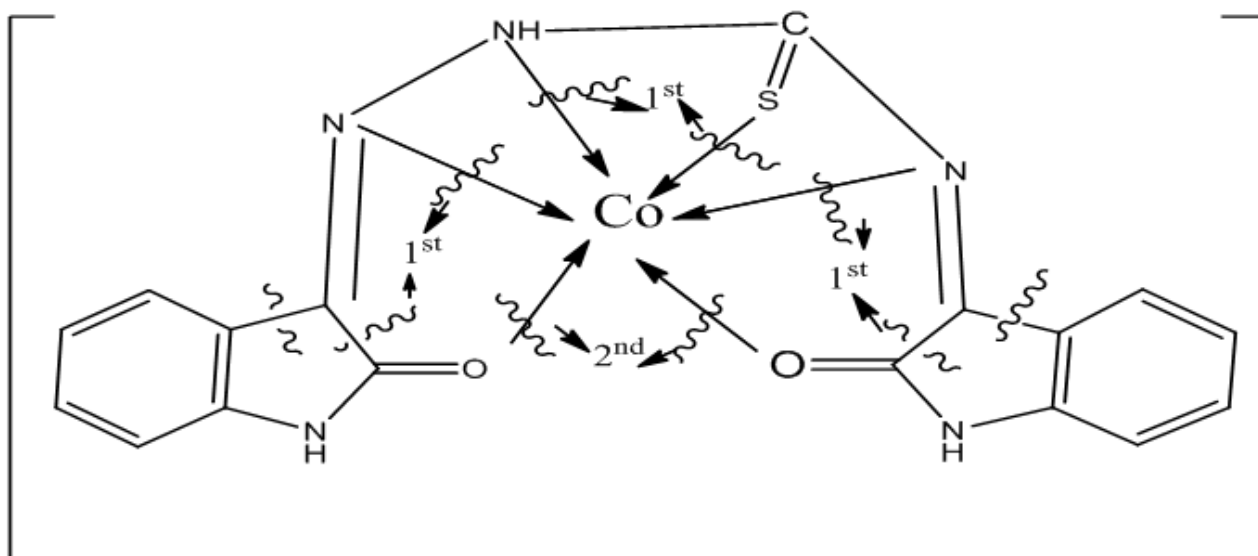


Fig. 10. Possible fragmentation pathway of  $[C_{17}H_{11}CoN_5O_2S]$ .

### 3.5. Antibacterial Activity

The prime objective of performing the antibacterial screening is to determine the susceptibility of the pathogenic microorganism. The tested compound which, in turn is used to selection of the compound as a therapeutic agent. The free Schiff base ligand and their metal complexes were screened for their antibacterial activity against strains the *Bacillus cereus* ATCC25923, *Escherichia coli* ATCC 25922, *Shigellasonnei*,

*Shigellaboydii*, *Enterobacter*, *Salmonella typhinium* of 4500. The compounds were tested at a concentration of 50 µg/ 0.01 mL in DMSO solution using the paper disc diffusion method [27-34]. The susceptibility zones were measured in diameter (mm) and the result were listed in table-5. The susceptibility zones were the clear zones around the discs killing the bacteria. All the Schiff base and metal complexes were individually exhibited varying degrees of inhibitory effects on the growth of tested bacterial species.

Table 5. Antibacterial screening activity of Schiff base and metal complexes.

Tested Bacteria	Diameter of zone inhibition(mm) of tested compounds				Kanamycin (30µg/disc)
	[SB]	Cr-com	Co-Com	Cd-com	
<i>Bacillus cereus</i>	7	8	11	10	20
<i>Bacillus subtilis</i>	8	9	10	13	28
<i>E. coli</i>	-	-	11	9	30
<i>Shigellasonnei</i>	8	10	12	-	27
<i>Shigellaboydii</i>	9	10	14	10	25
<i>Enerobacte</i>	-	10	8	8	21
<i>Salmonella typhinium</i>	8	10	13	-	30
DMSO control	-	-	-	-	30

Where, [SB]= Schiff base ligand[C<sub>17</sub>H<sub>11</sub>N<sub>5</sub>O<sub>2</sub>S]

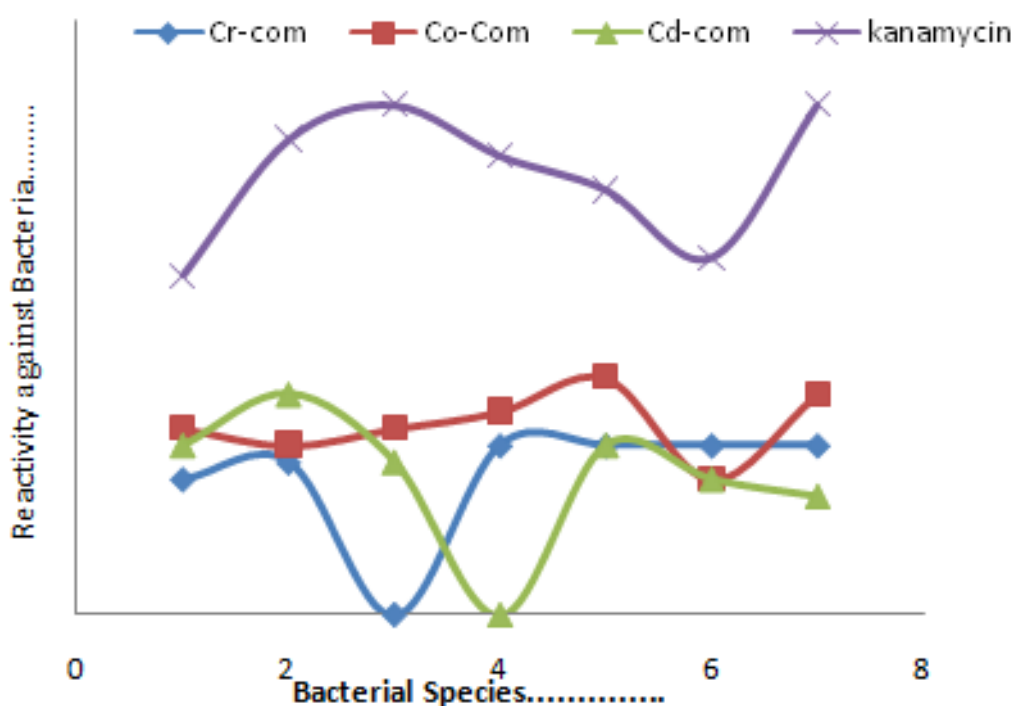


Fig. 11. Graphical representation of antibacterial screening effect of the tested complexes against seven pathogenic bacteria species.

## 4. Conclusion

In this paper we have explored the synthesis and coordination chemistry of Cr(III), Co(II) and Cd(II) complexes with new Schiff base ligand 2-bis(2-oxoindolin-3-ylidene) hydrazinecarbothioamide was derived from the condensation reaction of thiosemicarbazide and isatin. The physicochemical analysis indicated the formation of six coordinated metal complexes. IR spectral analysis indicated that N, O and S atoms were coordinated to central metal atom. Magnetic moment, UV-Visible and Thermogravimetric analysis confirmed the Proposed structure of Metal complexes. TGA analysis indicated that all the complexes are thermally stable up to 200°C and Cd(II) complex were more stable. Biological activity revealed that the ligand and its metal complexes have antibacterial activity as compared to the standard antibiotic (Kanamycin). The metal complexes had more antibacterial activity than its free Schiff base ligand.

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## References

- [1] B. Lakshmi, K. N. Shivananda, Gouda AvajiPrakash, Krishna Reddy K. Rama, and K. N. Mahendra,( 2011). Synthesis of Co(II), Ni(II) and Cu(II) Complexes from Schiff base Ligand and Reactivity Studies with Thermosetting Epoxy Resin. Bull. Korean Chem. Soc. 32 (5), 1613
- [2] Li-Yan Lai, Zheng Liu, Guo-Cheng Han, Zhencheng Chen., Springer, J Clust Sci. 2015, 26: 1845–1855.



- [3] Wilfredo Hernández, Juan Paz, Fernando Carrasco, Abraham Vaisberg, Evgenia Spodine, Jorge Manzur, Lothar Hennig, Joachim Sieler, Steffen Blaurock, and Lothar Beyer. Synthesis and Characterization of New Palladium(II) Thiosemicarbazone Complexes and Their Cytotoxic Activity against Various Human Tumor Cell Lines” Volume 2013, Article ID 524701, 12 pages.
- [4] Salman A. Khan, Abdullah M. Asiri, Khalid Al-Amry, and Maqsood Ahmad Malik “Synthesis, Characterization, Electrochemical Studies, and In Vitro Antibacterial Activity of Novel Thiosemicarbazone and Its Cu(II), Ni(II), and Co(II) Complexes” Volume 2014, Article ID 592375, 9 pages.
- [5] T. P. Yoon, E. N. Jacobsen, (2003). *Science* 299: 1691.
- [6] T. Yamada, T. Ikeno, Y. Ohtsuka, S. Kezuka, M. Sato, I. Iwakura, (2006). *Sci. Technol. Adv. Mater.* 7, 184.
- [7] Y. P. Cai, C. Y. Su, A. W. Xu, B. S. Kang, H. Q. Liu, S. Jie, (2001). *Polyhedron* 20, 657.
- [8] A. A. El-Asmy and G. A. A. Al-Hazmi, (2009). Synthesis and spectral feature of benzophenone-substituted thiosemicarbazones and their Ni(II) and Cu(II) complexes. *Spectrochimica Acta A*, 71 (5): 1885–1890.
- [9] T. A. Yousef, G. M. Abu El-Reash, O. A. El-Gammal, and R. A. Bedier, (2012). Co(II), Cu(II), Cd(II), Fe(III) and U(VI) complexes containing a NSNO donor ligand: synthesis, characterization, optical band gap, in vitro antimicrobial and DNA cleavage studies. *Journal of Molecular Structure*, 1029 (12): 149–159.
- [10] W. Liu, X. Li, Z. Li, M. Zhang, and M. Song, (2007). Voltammetric metal cation sensors based on ferrocenyl thiosemicarbazone. *Inorganic Chemistry Communications*. 10 (12): 1485–1488.
- [11] S.-M. Ying, (2012). Synthesis, crystal structure and nonlinear optical property of a Zinc(II) complex based on the reduced Schiff-base ligand. *Inorganic Chemistry Communications*, 22: 82-84.
- [12] C. Agatha Christia, C. Shijub and T. F. Abbs Fen Rejia. (2016). SYNTHESIS, SPECTROSCOPIC CHARACTERIZATION AND BIOLOGICAL ACTIVITIES OF CO(II), NI(II), CU(II) AND ZN(II) COMPLEXES OF SCHIFF BASE DERIVED FROM ISATIN MONOHYDRAZONE AND FURFURALDEHYDE” *jpmr*, 3 (20): 238-244.
- [13] Sridhar, S. K.; Pandeya, S. N.; Stables, J. P. and Ramesh, A. (2002). *Eur. J. Pharm. Sci.* 16: 129
- [14] Penthala, N. R.; Yerramreddy, T. R.; Madadi, N. R. and Crooks, P. A (2010). *Bioorg. Med. Chem. Lett.*, 20 (15): 4468
- [15] Pandeya, S. N.; Smitha, S.; Jyoti, M. and Sridhar, S. K. (2005) *Acta Pharm.*, 55: 27
- [16] Chiyanzu, I.; Hansell, E. and Gut, J. *Bioorg. (2003). Med. Chem. Lett.* 13: 3527.
- [17] Chohan, Z. H., Pervez, H. and Rauf, A. J. *Enz. Inhib. (2004). Med. Chem.*, 19: 417.
- [18] Md. Shiraj-U-Ddaula, Md. Anarul Islam, Shejutyaktar, Md. Khairul Islam, Md. Abdul Alim Al-Bari, Md. MasuqulHaque and Md. Kudrat-E-Zahan, 92014). Synthesis, Characterization and Antimicrobial Activity of Cd(II), Ni(II), Co(II) and Zr(IV) Metal Complexes of Schiff Base Ligand Derived from Diethylenetriamine and Isatin. *Asian J. Research Chem.* 7 (7): 619-621.
- [19] Bertrand, J. A.; Breece, J. L; Kalynaraman, A. R. Long, G. J. Bakes, W. A. J. *Am (1970). Chem. Soc.* 92: 5233.
- [20] Zahid H. Chohan, Asifa Munawar and Claudiu T. Supuran (2001). Transition metal complexes of some Schiff bases synthesis characterization and antibacterial studies. 8 (3).
- [21] Moamen S. Refat, H. K. Ibrahim, S. Z. A. Sowellim, M. H. Soliman, E. M. Saeed. (2009). Spectroscopic and Thermal Studies of Mn(II), Fe(III), Cr(III) and Zn(II) Complexes Derived from the Ligand Resulted by the Reaction Between 4-Acetyl Pyridine and Thiosemicarbazide. *Springer, J Inorg Organomet Polym* 19: 521–531.
- [22] Ahmed Jasim M. Al-Karawi, (2009). Synthesis and characterization of a new N2S2 Schiff base ligand and its complexes with nickel(II), copper(II) and cadmium(II) including the kinetics of complex formation. *Springer, Transition Met Chem* 34: 891–897.
- [23] Md. Saddam Hossain, Md. Ashraf Islam, C. M. Zakaria, M. M. Haque, Md. Abdul Mannan, Md. Kudrat-E-Zahan. (2016). Synthesis, Spectral and Thermal Characterization with Antimicrobial Studies on Mn(II), Fe(II), Co(II) and Sn(II) Complexes of Tridentate N,O Coordinating Novel Schiff Base Ligand” *J. Chem. Bio. Phy. Sci. Sec. A.*, 6 (1): 041-052.
- [24] M. R. Islam, J. A. Shampa, M. Kudrat-E-Zahan, M. M. Haque, Y. Reza., (2016). *J. Sci. Res.*, 8 (2), 181-189.
- [25] Md. Saddam Hossain, C. M. Zakaria, M. M. Haque and Md. Kudrat-E-Zahan, (2016) “Spectral and Thermal Characterization with Antimicrobial Activity on Cr(III) and Sn(II) Complexes containing N,O Donor Novel Schiff Base Ligand” *IJCS*, 4 (6): 08-11.
- [26] Alghool S, El-Halim HFA, El-sadek MA, Yahia I, Wahab L, (2015). Mononuclear complexes based on reduced Schiff base derived from L-methionine, synthesis, characterization, thermal and in vitro antimicrobial studies. *J Therm Anal Calorim.*, 121: 1309–1319.
- [27] N. Padma Priya, S Arunachalam, A. Manimaran. D. Muthipriya. C. Jayabalakrishnan *Spectrochim. Acta A* 72 (2009) 670-676.
- [28] M. R. Maurya, A. Kumar, M. Abid, and A. Azam, (2006). Dioxovanadium (V) and  $\mu$ -oxobis[oxovanadium(V)] complexes containing thiosemicarbazone based ONS donor set and their antiamoebic activity. *Inorganica Chimica Acta*, 359 (8): 2439–2447.
- [29] S. Padhye, A. Zahra, S. Ekk et al., (2005). Synthesis and characterization of copper(II) complexes of 4-alkyl/aryl- 1,2-naphthoquinones thiosemicarbazones derivatives as potent DNA cleaving agents. *Inorganica Chimica Acta*, 358 (6): 2023–2030.
- [30] R. P. Gupta and N. L. Narayana, (1997). Synthesis of some Mannich bases of 1-cyclohexylidene-N (1,2-dihydro-2-oxo-3H-indol-3-ylidene) thiosemicarbazones and their antibacterial activity, *Pharmaceutica Acta Helvetica*, 72 (1): 43–45.

- [31] T. A. Yousef, G. M. Abu El-Reash, O. A. El-Gammal, and R. A. Bedier, (2012). Co(II), Cu(II), Cd(II), Fe(III) and U(VI) complexes containing a NSNO donor ligand: synthesis, characterization, optical band gap, in vitro antimicrobial and DNA cleavage studies. *Journal of Molecular Structure*, 1029 (12): 149–159.
- [32] Moamen S. Refat, H. K. Ibrahim, S. Z. A. Sowellim, M. H. Soliman, E. M. Saeed, (2009) Spectroscopic and Thermal Studies of Mn(II), Fe(III), Cr(III) and Zn(II) Complexes Derived from the Ligand Resulted by the Reaction Between 4-Acetyl Pyridine and Thiosemicarbazide. Springer, *J InorgOrganomet Polym.*, 19: 521–531.
- [33] Xinde Zhu, Chenggang Wang, Zhiping Lu and Yuanlin Dang, (1997). Synthesis, characterization and biological activity of the Schiff base derived from 3, 4-dihydroxybenzaldehyde and thiosemicarbazide, and its complexes with nickel(II) and iron(II). Springer, *Transition Met. Chem.*, 22: 9-13.
- [34] T. Rosu, A. Gulea, A. Nicolae, and R. Georgescu, (2007). Complexes of 3dn metal ions with thiosemicarbazones: synthesis and Bioinorganic Chemistry and Applications 11 antimicrobial activity. *Molecules*, 12 (4): 782–796.