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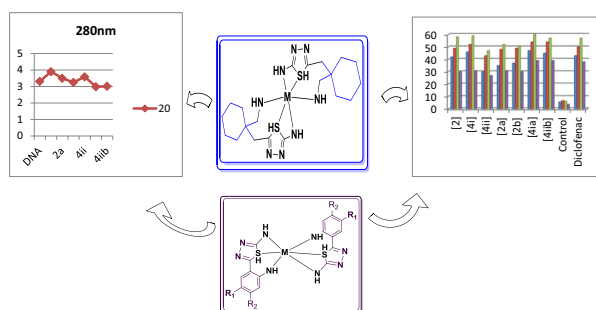
## DNA binding, anti-inflammatory and analgesic evaluation of metal complexes of N/S/O donor ligands; Synthesis, spectral characterization

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### HIGHLIGHTS

- Effective biological compounds like organic ligands were synthesized.
- Synthesized various biological active metal complexes.
- Systematically characterized by various physico-chemical analysis of ligands and metal complexes.
- The excellent analgesic and anti-inflammatory activity carried out for synthesized compounds.
- We report a few metal complexes of highly anti-inflammatory agents.

### GRAPHICAL ABSTRACT



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### ABSTRACT

Transition metal complexes containing tri-dentate NSN donor ligands i.e., 5-((1(aminomethyl)cyclohexyl)methyl)-1,3,4-thiadiazol-2-amine (AMTA) (**2**) and 5-(2-aminophenyl)-1,3,4-thiadiazol-2-amine (ATA) (**4i–ii**) have been synthesized. The newly synthesized ligands and their respective complexes were characterized by elemental analysis, molar conductance measurement and various spectral studies [infrared (IR), electronic, and NMR (for ligands only)]. Metal complexes are like  $[M(AMTA)_2]$ ,  $[M(ATA)_2]$  type, where M = Mn(II), Co(II) and Cu(II). The proposed geometries of the complexes are octahedral in nature. The synthesized ligands and their complexes exhibit effective anti-inflammatory, analgesic and DNA binding activities. All the tested compounds exhibited significant analgesic activity, whereas the compound **4i**, **4(ia)** and **4(iib)** is equipotent with Diclofenac sodium.

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### Introduction

Ruthenium(II) polypyridine complexes have been the subject of intense interest from different perspectives for DNA binding studies [1–4]. Copper complexes of several ligands have been prepared

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and evaluated for anti-inflammatory activity. Other compounds known for their anti-inflammatory properties are the S, N-heterocyclic ligands, e.g. thiazoline and its derivatives [5–10]. The synthesis of new coordination compounds containing nitrogen, oxygen and sulfur donor atoms has been significantly developed. Among these materials, thiosemicarbazide compounds are specifically considered by researchers because of biological properties such as antibacterial [11], antimalarial [12], antiviral [13], antitumor [14,15], insecticidal [16] and optoelectronic properties such as good non-linear optical (NLO) response [17]. Actually, thiosemicarbazide constitute one of the most important classes of mixed hard-soft NSO donor ligands [18,19]. A change in the substitution

pattern of these compounds can create new ligands with different properties.

Gabapentin (neurontin) (GBP) is a neuroleptic drug, and is important because of its biological significance and selectivity towards the oxidant. It is sometimes prescribed for the management of neuralgia (nerve pain) [20]. Gabapentin has been prescribed off-label for the treatment of some mood disorders, anxiety and tardive dyskinesia (a neurological syndrome caused by the long-term use of neuroleptic drugs). Diperoxidatonickelate(IV) (DPN) as an oxidant in alkaline medium is new and restricted to a few cases [21–23] due to the fact of its limited solubility and stability in aqueous medium. Reduction of nickel(IV) complexes have been received a considerable attention in order to understand, the nature of intermediate oxidation states of nickel such as nickel(III). Indeed, stable nickel(III) complexes are known [24,25].

Herein, we report the series of synthesized complexes,  $[M(L^1)_2]$  (**2a–c**) and  $[M(L^2)_2]$  (**4ia–c**) (**4iia–c**) by using synthetic strategy reported earlier with slight modification. The DNA binding activity of the metal complexes was performed using agarose gel electrophoresis technique. The synthesized compounds were also tested for their analgesic and anti-inflammatory activities. The objective of the current study is to provide useful insights in the understanding of analgesic and anti-inflammatory potency of transition metal complexes.

## Results and discussion

The physical and analytical data of the ligands and their corresponding complexes are depicted in (Table 1).

### Infrared spectroscopy

IR spectra of the new metal complexes reveals the following points:

- The spectra in the region  $3125\text{--}3350\text{ cm}^{-1}$  is due the presence of  $\text{--NH}$  functional group in free ligand, which leads the shifting of frequency to lower region of about  $50\text{--}75\text{ cm}^{-1}$ , indicates that the formation of complexes through amino group.
- A sharp band appear in the region  $1600\text{--}1670\text{ cm}^{-1}$  in ligand which is represents the presence of azomethane ( $\text{--C=N}$ ) group in thiadiazole ring of ligands. This band frequency was unaltered in complexes.
- IR data supports the involvement of the amino nitrogen in coordination of metal ions at  $450\text{--}520\text{ cm}^{-1}$ . The spectra shows a band at  $550\text{--}620\text{ cm}^{-1}$  supports the involvement of sulfur atom in coordination with metal ions [32,33].

### Molar conductance studies

The molar conductivity values in the range  $15.25\text{--}22.00\text{ ohm}^{-1}\text{ cm}^2\text{ mol}^{-1}$  in DMSO denote that all the complexes behave as 1:2 electrolytes in these solvents [34] and all metal complexes shows non-electrolytic characteristic property.

### Electronic spectra

Electronic spectra of the ligands are shown in the (Figs. 2 and 3) and the metal complexes Co(II) and Cu(II) in the (Figs. 4 and 5), these spectra's were depicted that the bands below  $\sim 340\text{ nm}$  are attributed to intraligand transition [35]. A small shift should be observed for the second band in all complexes, these  $\pi \rightarrow \pi^*$  transitions probably involving metal and ligand orbital's. Bands above  $\sim 340\text{ nm}$  are ascribed to charge transfer process, probably from ligand to metal and mainly associated with the amine and acidic

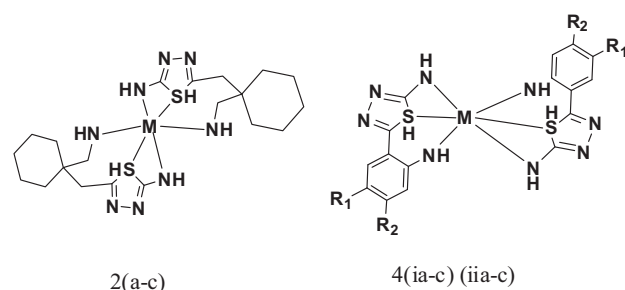


Fig. 1. Tentative geometry of metal complexes.

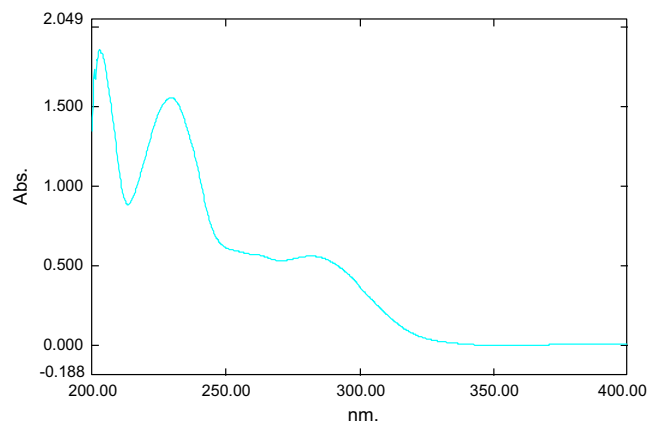


Fig. 2. UV-VIS spectra of ligand (2).

Table 1

Molecular formulae, color, molar conductance and magnetic susceptibility data of the ligand and its metal complexes.

Compounds	Mol. formulae	Color	Molar conduct. ( $\text{Ohm}^{-1}\text{ cm}^2\text{ mol}^{-1}$ )	$\mu_{\text{eff}}$ (BM)
[2]	$\text{C}_{10}\text{H}_{18}\text{N}_4\text{S}$	White	–	–
[4i]	$\text{C}_8\text{H}_7\text{N}_5\text{O}_2\text{S}$	White	–	–
[4ii]	$\text{C}_8\text{H}_7\text{ClN}_4\text{S}$	White	–	–
[2a]	$\text{C}_{21}\text{H}_{37}\text{MnN}_8\text{S}_2$	Brownish	79.12	5.71
[2b]	$\text{C}_{21}\text{H}_{37}\text{CoN}_8\text{S}_2$	Green	84.15	3.65
[2c]	$\text{C}_{21}\text{H}_{37}\text{CuN}_8\text{S}_2$	Bluish	89.17	1.19
[4ia]	$\text{C}_{16}\text{H}_{12}\text{MnN}_{10}\text{O}_4\text{S}_2$	White	76.51	5.61
[4ib]	$\text{C}_{16}\text{H}_{12}\text{CoN}_{10}\text{O}_4\text{S}_2$	Green	86.25	3.66
[4ic]	$\text{C}_{16}\text{H}_{12}\text{CuN}_{10}\text{O}_4\text{S}_2$	Green	91.04	1.21
[4iia]	$\text{C}_{16}\text{H}_{12}\text{Cl}_2\text{MnN}_8\text{S}_2$	Grey	71.51	5.74
[4iib]	$\text{C}_{16}\text{H}_{12}\text{Cl}_2\text{CoN}_8\text{S}_2$	Bluish	82.25	3.25
[4iic]	$\text{C}_{16}\text{H}_{12}\text{Cl}_2\text{CuN}_8\text{S}_2$	Brownish	77.04	1.21

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