Life Science Informatics Publications

Research Journal of Life Sciences, Bioinformatics, Pharmaceutical and Chemical Sciences

Journal Home page http://www.rjlbpcs.com/

RJLBPCS

ISSN 2454-6348

Original Review Article **DOI: 10.26479/2018.0405.56**

A REVIEW: LANTHANIDE COMPLEXES AND THEIR BIOLOGICAL IMPORTANCE

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ABSTRACT: The present review stems from our interest in biological property of lanthanide complexes derived with different kinds of ligands or organic compounds. Lots of research on lanthanide metals and their complexes have been done and numerous research articles are published till to day. Lanthanide ions and their compounds received the industrial and academic attention due to their great medicinal importance. Lots of biologically important organic compounds are reported as more biologically active compounds on complexation with lanthanide metals/ions. From many research articles it has been reported that lanthanide metals with various organic compounds can act as optimum antimicrobial agents as well as various kinds of therapeutic agents also.

KEYWORDS: Lanthanide complexes, biological importance, therapeutic agents.

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1.INTRODUCTION

Coordination chemistry of lanthanide metals is one of the impulsive research field in inorganic chemistry [1]. It appears from the investigation of reported literature that lanthanides are competent to form stable complexes with various Schiff bases and many other newly synthesized compounds or its derivatives [2]. It has been also found for the lanthanide complexes that they exhibit the anticancer, antibacterial, antifungal and many other variable medicinal properties [3-6]. Compounds of gadolinium are widely used as magnetic resonance imaging (MRI) agents [7]. The biological activities of the lanthanides with coumarin and its related compounds show the photobiological properties, antitumor, anti-leukemia and anti-HIV activities [8-11] while heterocyclic compounds

of lanthanides express agricultural properties. It has been also found that trivalent lanthanide-nitrogen donor compounds are applicable in catalysis and organic synthesis [12,13].

The Biological importance of lanthanide complexes

Zhang et al. reported the four lanthanide complexes by using lanthanide ion and NITph-p-Cl radical (nitronyl nitroxide) with hexafluoro acetylacetonate (hfac) ligand. $Ln(hfac)_3(NITph\text{-}p\text{-}Cl)_2$ types of complexes were synthesized using Er, Tb, Dy and Gd metals and tested them for antibacterial activity against some bacterial species. The study of antibacterial activity showed that all Ln complexes revealed the better antibacterial activity than lanthanide ions and radicals. It has been proved from the antibacterial study that the complex of Gd exhibited the optimal antibacterial activity against *Escherichia coli* than that of other three Ln complexes. Other three complexes of Er, Tb and Dy showed good antibacterial activity against *Escherichia coli* but they were reported weaker than that of Gd. The Gd complex can be used as antibacterial agent for various kind of applications due to its best antibacterial property [14]. Z. A. Taha et al. synthesized complexes of trivalent La, Gd, Nd, Pr, Er, Sm, Tb and Dy metals with Schiff base ligand (N,N-bis(1-naphthaldimine) which was derived from 2-hydroxy-1-naphthaldehyde and o-phenylene diamine. The ligand contains two phenolic oxygen atoms and two nitrogen atoms of imine for coordination with central metal. Biological features of ligand and its Ln(III) complexes against some bacteria revealed that ligand against *p. aeruginosa* and *E.coli* bacteria showed ordinary antimicrobial activity and did not showed activity against *p. vularis*, *s. dysenteriae*, *s. aureus*, Klebsiella & Serratia while synthesized La and Pr complexes showed the highest antimicrobial activity against *s. aureus* than the complex of Gd, Nd, Sm, Tb, Er and Dy, those does not possessed good antimicrobial activity against *s. aureus.* All lanthanide complexes with (N,N-bis(1-naphthaldimine) ligand indicated higher antimicrobial activity against gram-positive bacteria while they showed poor antimicrobial activity against gram-negative bacteria [15]. R. Gupta et al. synthesized binary complexes of trivalent La, Sm, Gd and Dy using FCA (Furan-2-carboxylic acid) as a ligand. The Antibacterial and Antifungal activities of the complexes was tested against some gram negative and gram-positive bacterial species. From the antibacterial and antifungal study synthesized complexes showed more prohibitive activity against some bacteria and fungi in the comparison of parent ligand. It was studied that the binary complexes of La, Sm, Gd and Dy with FCA ligand revealed remarkable antifungal activity against *Aspergillus niger*, *Aspergillus fumigatus* and *Aspergillus flavus* and antibacterial activity against *Escherichia coli* (gram negative) and *Staphylococcus aureus* (gram positive). While $Eu(tfn)_3$(phenedione), $Eu(hft)_3$(phenedione) and $Yb(hfa)_3$(phenedione) complexes were derived by M. Abdus Subhan et al. from the reaction of $Eu(tfn)_32H_2O$, $Eu(hft)_32H_2O$ and $Yb(hfa)_32H_2O$ compounds with phenedione respectively. Structural properties of complexes were characterized by PL spectra (Photo Luminescence spectra) as well as by other spectral techniques and the massive antibacterial activity of Phenedione and its lanthanide complexes was established

from the antibacterial study and exposed the highest antibacterial activity against *p. penneri* bacteria than another two bacterial species *Escherichia coli* and *Staphylococcus aureus* [16,17]. The Antimicrobial study of trivalent lanthanide metal complexes was reported by Waleed Mahmoud Al Momani et al. The complex series of trivalent lanthanide metals with Schiff base ligand was derived from 2-hydroxy-1-naphthaldehyde with o-phenylenediamine and it was proved from the antimicrobial activity that most of Ln complexes revealed the higher activity against some bacteria than the free ligand and the complexes of La and Pr were found to more active against *P. aeruginosa* than the standard antibiotics like cephalexin and cephradine and it was also found that the gram-positive bacteria were much more sensitive than the gram-negative bacteria towards the Ln complexes. P. Kapoor et al. reported the two octahedral complex series of Nd, Sm and Gd metals using two ligands 3-acetylcoumarin thiosemicarbazone (ACTSZH) and 3-acetyl coumarin semicarbazone (ACSZH) **(Fig.-1)** one by one. These two series of lanthanide complexes synthesized by microwave and thermal methods.

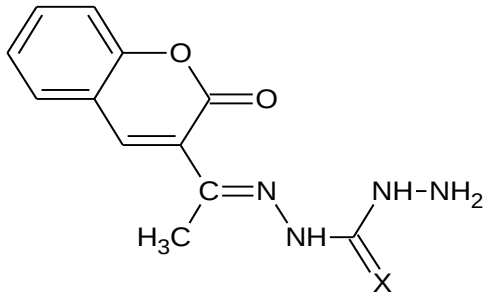

Fig.-1: 3-acetylcoumarin thiosemicarbazone (X= S-atom),
3-acetyl coumarin semicarbazone (X= O-atom)

Determination of the antibacterial and antifungal activities of ligands and complexes were carried out against some bacterial and fungal strains and substantiated the good biological activity of Ln complexes than their uncomplexed ligands and the complexes of Sm with both ligands (ACTSZH and ACSZH) were found to have the highest biological activity than the other complexes and parent ligands [18,19]. A series of lanthanide complexes were reported by G. Karthikeyan et al. with tetracycline hydrochloride (TC) ligand using the salt of trivalent La, Pr, Nd, Sm, Gd, Tb, Dy and Y. The antibacterial activity of ligand and its complexes was studied on gram-negative and gram-positive bacterial species which substantiated the more toxic nature of these lanthanide complexes against gram positive bacterial species-*Staphylococcus aureus* than gram negative bacteria named *Escherichia coli* while TC ligand has been tested less toxic than its complexes against *Staphylococcus aureus*. So, it was absolute that these complexes were produced bacterial growth inhibition against the gram-positive bacteria while ligand showed good antibacterial activity than its complexes against *Escherichia coli* that means the complexes of lanthanide metals with TC ligand showed less antibacterial activity than parent ligand against gram negative bacteria (*Escherichia coli*) while complexes showed good antibacterial activity than its parent ligand against

gram positive bacteria [20]. L. Lekha et al. reported the biological study of such trivalent lanthanide complexes synthesized with the Schiff base ligand named N-[(Z)-(5-bromo-2-hydroxyphenyl)methylidene]-2-hydroxyacetamide (**Fig.-2)** was derived from L-Serine and 5-bromo salicylaldehyde.

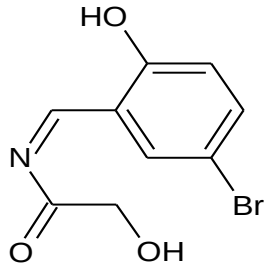

Fig.-2: N-[(Z)-(5-bromo-2-hydroxyphenyl)methylidene]-2-hydroxyacetamide

It was proved from the biological study that the Ln(III) complexes contain good antibacterial properties than the free Schiff base ligand against *E. coli*, *P. vulgaris*, *P. aeruginosa* and *S. aureus* bacteria. The Schiff base ligand was biologically active due to its imine group but on the complexation with such lanthanide metals exhibit the higher antibacterial activity than the free Schiff base ligand. The Gd and Sm Schiff base complexes showed considerable enhancement in antibacterial activity than the other Ln complexes and its uncomplexed ligand [21]. Mohanan and Devi synthesized the two series of novel lanthanide complexes using chloride and nitrate salts of lanthanide metals with 2-(N-Salicylidene amino)-3-carboxyethyl-4,5,6,7-tetrahydro benzo[b]thiophene (HSAT) ligand **(Fig.-3)** derived by 2-amino-3-carboxyethyl-4,5,6,7-tetrahydro benzo[b]thiophene with salicylaldehyde.

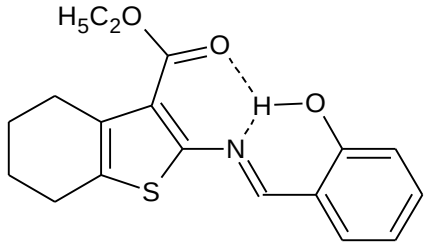

Fig.-3: 2-(N-Salicylidene amino)-3-carboxyethyl-4,5,6,7-tetrahydro benzo[b]thiophene

These two series of the novel complexes with coordination no. 12 were tested for biological property. From the 20 complexes of La, Pr, Nd, Sm, Eu, Gd, Dy, Tm, Yb and Lu metal six of La, Nd and Sm were selected for the biological test. The antibacterial and antifungal activities of ligand and its six selected complexes against some pathogenic bacterial and fungal species were tested and established that the ligand was physiologically active while its complexes elongated their biological activities. Complexes of La, Nd and Sm found to have good antibacterial and antifungal properties better than ligand against some pathogenic bacteria and fungi species. A. Moodi et al. studied the binding analysis between Yb type of complex and DNA. The trivalent Yb complex containing 1,10-phenanthroline (phen). The binding analysis was investigated of the complex with DNA by

absorption, fluorescence spectroscopy, gel electrophoresis and viscosity measurement. A study suggested the interaction mode between Yb complex and DNA to be groove binding. The antimicrobial activity was also reported for this complex against gram positive (*M. luteus*, *B. cereus*, *Bacillus*) and gram negative (*S. marcescens*, *shigella*, *A. baumannii*, *K. pneumoniae*, *S. paratyphi C*, *S. paratyphi B*, *E. coli*, *Enterococcus*) bacterial species and it was revealed that the Yb complex is strongly active as an antibacterial agent [22,23]. Lanthanide complexes of trivalent Eu, Gd, Nd, Sm and Tb with Phenylthioacetic acid (PTAA) reported by T.F. Abbs Fen Reji et al. Their antimicrobial activity, DNA cleavage analysis and anticancer activity were also reported. DNA cleavage study indicated that the Eu and Nd complexes cleaved the DNA completely than other complexes as well as compounds except free ligand were found to show better antibacterial activity rather than antifungal activity while Nd complex showed the higher antifungal activity than other metal complexes and also found the good activity against gram negative species of bacteria such as *E. coli* and *B. subtilis*. The study of anticancer activity of ligand (PTAA) and its lanthanide complexes on cancer cells such as HeLa (Human cervical cancer cells) and HCT116 (colon cancer cells) substantiated the most inhibitory effect. Eu and Nd complexes found to possess the strongest inhibitory effect on the growth of the both cancer cells in the comparison of other three complexes and its free ligand So, it was reported that complexes of Nd and Eu with PTAA ligand were more active on the both cancer cell, HeLa and HCT116. Trivalent lanthanide complexes of La, Pr, Nd, Sm and Ho with Phenylthiopropionic acid (PTPA) ligand were synthesized and studied for medicinal properties by C. Shiju et al. Characterizations of these compounds were made on the basis of thermal analysis, molar conductance, powder XRD, IR, mass and electronic spectra. The important studies like DNA cleavage, In vitro anticancer activity and antimicrobial activity were carried out for these compounds and it was conformed from the study of DNA cleavage that complexes of Pr and Nd completely cleaved the DNA than other complexes. One of the important studies of In vitro anticancer activity it was absolutely revealed that the La and Nd complexes showed the most inhibitory effect on the growth of HeLa (Human cervical cancer cells) and HCT116 (Colon cancer cells) like cancer cells than the complexes of Pr, Sm, Ho and its uncomplexed ligand while the good antibacterial and antifungal properties were reported for these complexes. A better activity was found for the bacteria rather than the fungi but only Nd complex was found to exhibit the higher activity against fungi and gram-negative bacteria such as *E. coli* in the comparison of other complexes and free ligand [24,25]. Newly mixed ligand metal complexes of lanthanide metals with mixed bivalent N, O donor Schiff base ligand named 2-(E)-[4-methoxybenzylidene-amine]phenol **(Fig.-4)** and 2-(E)-[4-methoxyphenylimino methyl]-6-methoxyphenol **(Fig.-5)** synthesized by V.D. Ingale et al. Schiff bases were derived from Salicylaldehyde with 4-methoxyphenylamine and 2-hydroxy-3-methoxybenzaldehyde with 4-methoxyphenylimine respectively.

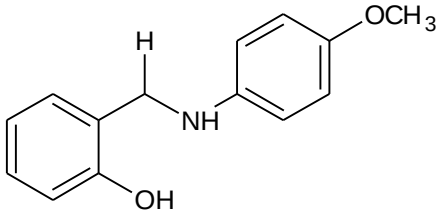

Fig.-4: 2-(E)-[4-methoxybenzylidene-amine]phenol

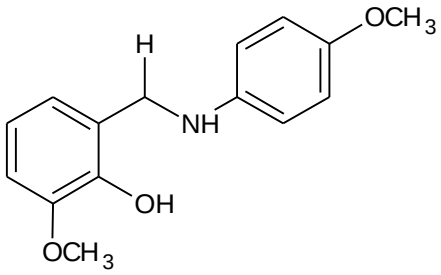

Fig.-5: 2-(E)-[4-methoxyphenylimino methyl]-6-methoxyphenol

Antibacterial activity of the lanthanide complexes tested against *staphylococcus aureus* and *bacillus substilis* while antifungal activity was tested against *aspergillus niger* and *fusarium oxysporum* and it was exposed that these complexes were highly biological active than uncomplexed ligand while the mixed ligands Tb complex found to have good antibacterial activity in comparison of the standard drugs ciprofloxacin and other complexes as well as ligand and complexes of Ce and Gd were found to substantiated the antifungal activity higher than other complexes and its parent ligands. Lanthanide complexes were derived by S.T. Gaikwad et al. using 2-(E)-(4-methoxyphenylimino)methyl phenol (MPMP) divalent Schiff base ligand. The new ligand 2-{[(4-methoxyphenyl)amino]methyl}phenol **(Fig.-6)** was derived from salicylaldehyde and 4-methoxyaniline. The biological properties were tested against *staphylococcus aureus* and *bacillus substilis* (gram positive) pathogens.

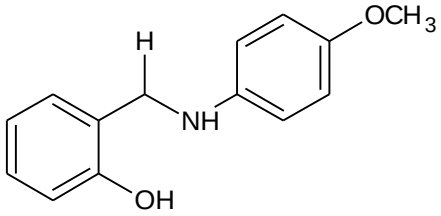

Fig.-6: 2-{[(4-methoxyphenyl)amino]methyl}phenol

It was proved from the biological study that Gd complex exposed the higher antibacterial activity than other and it was enhanced in the activity against the species *staphylococcus aureus* while study of antifungal activity against the fungal species *aspergillus niger* and *fusarium oxysporum* (gram negative) suggested that the Gd complex against *fusarium oxysporum* and La complex against *aspergillus niger* species showed higher antifungal activities than other complexes [26,27]. Preeti Singhal et al. reported the antimicrobial study of their newly synthesized complexes of trivalent

lanthanide metals with the ligand Cefepime. Antimicrobial study of the cefepime ligand and its trivalent Ln complexes have been extensively studied against one gram negative- *Escherichia coli* and two gram positive- *Staphylococcus aureus* and *Streptococcus pyogenes* types of bacterial strains. The antibacterial study for cefepime suggested that the cefepime showed the strong activity against the gram-positive bacteria in the comparison of the gram-negative bacteria while all lanthanide complexes were found to be more active against the *Escherichia coli* and *Staphylococcus aureus* than its free ligand but the Pr complex showed the highest activity against *Escherichia coli* (gram positive) and *Staphylococcus aureus* (gram negative) compared to the other Ln-complexes. So, it was clear that the complexation enhanced the antimicrobial activity in the comparison of free ligand [28]. Newly eight Ln-complexes of N, N'-bis(2-hydroxynaphthylmethylidine)-o-phenylenediamine Schiff base ligand **(Fig.-7)**. Biological properties of the complexes were established against three gram-negative bacterial species.

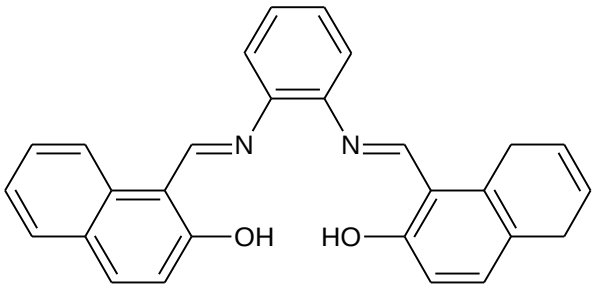

Fig.-7: N, N'-bis(2-hydroxynaphthylmethylidine)-o-phenylenediamine

It has been established from the biological activity against pathogenic bacterial species that the ligand was inactive against *s. dysenteriae* and *p. vulgaris* but it showed moderate activity against *p. aeruginosa* while only Sm complex was highly active against *p. aeruginosa* in compared to other complexes and from all complexes only Sm and Dy complexes expressed the high activity against *s. dysenteriae* and the complexes of Pr and Dy exhibited the higher activity against *p. vulgaris* than others. From the two complexes of trivalent La and Sm with enrofloxacin (ER) ligand named 1-cyclopropyl-7-(4-ethylpiperazin-1-yl)-6-fluoro-4-oxo-1,4-dihydroquinoline-3-carboxylic acid **(Fig.-8)**, the good antibacterial property of Sm complex was reported by Yan-Jun Wang et al.

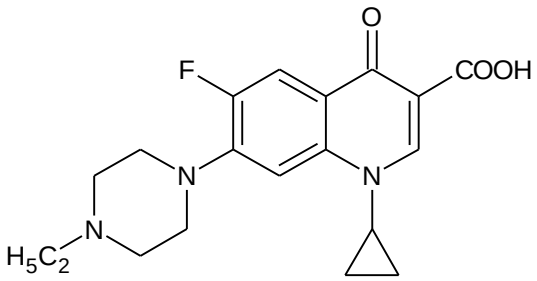

Fig.-8: 1-cyclopropyl-7-(4-ethylpiperazin-1-yl)-6-fluoro-4-oxo-1,4-dihydroquinoline-3-carboxylic acid

Both complexes were biologically tested against gram positive (*Bacillus subtilis*, *Staphylococcus aureus*) and gram negative (*Escherichia coli*) bacterial species which substantiated that the complexation enhanced the antibacterial activity and it was found that the Sm complex exhibited the highest antibacterial activity than La complex and its uncomplexed ligand [29,30]. Lanthanide metal complexes of PPCADQ (2-pyridine-2-yl-3-[pyridine-2-carboxylideneamino]-1,2-dihydroquinazoline-4-(3H)-one) were reported as moderate biological active compounds by Gudasi et al. The ligand and its complexes were screened for the antibacterial and antifungal activity and from the study it has been reported that PPCADQ ligand was moderate active against *Aspergillus niger* while it was less active against *Penicillium notation*, *Pseudomonas aeruginosa* and *Bacillus cirroflagellosus*. The complexes were moderately activity against *Pseudomonas aeruginosa* in comparison of the ligand. Complexes of trivalent Sm, Eu, Gd, Tb, Dy and Y exposed the moderate activity against *Bacillus cirroflagellosus* but the complexes of trivalent La, Pr and Nd were found to be less active against *Bacillus cirroflagellosus* and it was found that the complexation enhanced the antibacterial activity. The synthesis of the ternary complexes of Cerium with 2,3-dimethyl-1-phenyl-4-salisaldehydene-3-pyrazoline-5-one Schiff base and five amino acids as a ligand and the study of antibacterial property were reported by B.D. Aghav et al. The antibacterial behavior was tested for the cerium complexes against bacterial species viz. *S. aureus*, *C. diphtheriae*, *P. aeruginosa* and *E. Coli* was exposed that all complexes exhibited the outstanding antibacterial activity than each ligand against *S. aureus*, *C. diphtheriae*, *P. aeruginosa* and *E. Coli* and as reported it has been obviously concluded that all these Ce complexes can be good antibacterial agents especially against *E. coli* than the other bacterial species [31,32].

2. CONCLUSION

Synthesis of lanthanide metal complexes with different kinds of organic compounds is becoming an extensive interesting area for the researchers because of their massive pharmacological importance. Metal complexes of lanthanides are preferred due to coordination with different chelators or ligand systems showing potential biological properties. Up to now, the main focus to synthesize the promising biologically important agents is still around the alteration and optimization of Ln complexes by using as many ligands as possible which express the outstanding biological features. This review show that the complexes of Ce, Gd and Nd show extensive antifungal activity while Gd complex with particular ligand (hexafluoro acetylacetonate) can also be used as a potent antibacterial agent. Complexes of Yb, Eu, Nd and few other lanthanide metals found to performed as anticancer agent against particular cancer cell. It has been concluded from this review that lanthanide complexes with particular organic compound(s) can contributing the best in the development of new therapeutic agents. This review can be helpful to develop the drug/medicine for particular diseases caused by particular bacterial and/or fungal species.

CONFLICT OF INTEREST

The authors have declared that they have no conflict of interest.

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