Est. 1984

ORIENTAL JOURNAL OF CHEMISTRY
An International Open Access, Peer Reviewed Research Journal
www.orientjchem.org

ISSN: 0970-020 X
CODEN: OJCHEG
2021, Vol. 37, No.(3):
Pg. 575-582

Neodymium (III) and Samarium (III) Complexes of 2, 2′-((1E, 1′E)-(1, 2- Phenylene bis(Azanlylidene)bis(Methanylylidene)) diphenol ligand: Synthesis, Characterization and Biological activity

EMAIME JIMMY UWANTA[1]*, ATIM SUNDAY JOHNSON[2], PIUS OZIRI UKOHA[3] and EMMANUEL JIMMY UKPONG[4]

[1,4] Department of Chemistry, Faculty of Physical Sciences, Akwa Ibom State University, Ikot Akpaden. Akwa Ibom State. Nigeria.
[2]Department of Chemistry, Faculty of Sciences. University of Uyo, Uyo. Akwa Ibom State. Nigeria.
[3]Department of Pure and Industrial Chemistry. Faculty of Physical Sciences, University of Nigeria, Nsukka, Enugu State, Nigeria.
*Corresponding author E-mail; emaimeuwanta@gmail.com

http://dx.doi.org/10.13005/ojc/370308

(Received: April 06, 2021; Accepted: June 18, 2021)

ABSTRACT

Complexes of Nd(III) and Sm(III) of a Schiff base ligand derived from the condensation of *o*-phenylenediamie and 2-hydroxybenzaldehyde were synthesised and characterised by various analytical and spectroscopic techniques. The analytical values revealed a metal to ligand ratio of 1:2 for the Nd(III) complex and a 1:1(ligand: metal) for Sm(III) complex. The complexes were characterised by elemental analysis, molar conductance, UV–Visible and infrared spectral studies. Plausible structures were proposed for Nd(BSOPD)$_2$ and SmBSOPD based on the results obtained. *In-vitro* antimicrobial activities of both the Schiff base and the synthesised metal complexes were evaluated against six (6) human pathogens. The complexes showed significant activity compared to the free ligand. These complexes may be employed as active ingredients in the development of antibacterial and antifungal agents.

Keywords: Schiff Base, Bis(salicylaldehyde)orthophenylenediamine, Metal Complexes, Biological activity.

INTRODUCTION

The lanthanides also called rare earth elements are found between lanthanum and lutetium of the Periodic Table. These elements have a greater propensity towards the formation of electrostatic linkages rather than covalent bonds as shown in their structural chemistry; lanthanide (III) ions may form complex compounds possessing higher coordination numbers with monodentate, simple bidentate or polydentate ligands.[1] The structural flexibility is due to the absence of strong crystal efforts for the 4f electronic configurations in addition to the bulky ionic radii of these lanthanide ions, which change significantly with atomic number or oxidation state of the lanthanides.

Lanthanide metals and their compounds are distinctly unique due to their spectroscopic, physical, and chemical characteristics. The lanthanide complexes have been found to exhibit anticancer, antibacterial, antifungal and many other variable medicinal properties[2-5]. Lanthanide complexes have been successfully applied as diagnostic tools in biomedical examination such as magneticr imaging. Compounds of gadolinium, for example, are widely used as contrast agents in magnetic resonance imaging(MRI)[6]. The lanthanides also, exhibit noticeable biological activity as some are capable of replacing Ca^{2+} ion in organised molecules[7-9]. The biological activities of the lanthanides with coumarin and its related compounds show the photobiological properties, antitumor, anti-leukemia and anti-HIV activities[10-13]. One of the essential elements of the lanthanides is samarium and its ion, Sm^{3+} finds application in high-density optical storage and antibacterial properties[14-15]. A Schiff base is formed when there is a condensation reaction between an amine and an aldehyde or ketone. They are readily applied in spectrophotometry, catalysis, medicine, metallurgy and magnetic materials. Schiff bases are important in the advancement of coordination chemistry as they form stable complexes easily with transition metals. Analysis of reported literature reveals that lanthanide elements are capable of forming stable complexes with various Schiff bases and many other synthesized compounds or their derivatives[8]. Metal complexes of Mg(II), Zn(II), Sn(II), VO(II) and Bi(III) ions containing the Schiff base ligand {2,2`((1E,1`E)-(1,2-phenylenebis(azanylylidene)) (methanylidene)) diphenol} have been reported[16]. However, to the best of our knowledge, no lanthanide metal complexes with the Schiff base ligand; 2,2′-((1E, 1′E)-(1, 2-phenylene-bis(azanlylidene)-bis(methanylylidene))diphenol (BSOPD) and their biological activity are described. Therefore, in this paper, the synthesis, characterization and antimicrobial study of neodymium (III) and samarium (III) complexes using the Schiff base BSOPD is reported.

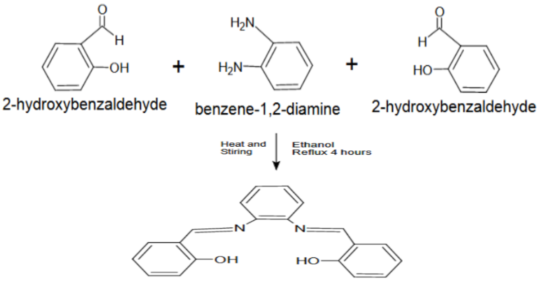
MATERIALS AND METHOD

Analytical grade reagents were used throughout this study. Neodymium(III)nitrate.6H_2O, samarium(III)chloride and 1,2-diaminobenzene were purchased from Sigma- Aldrich.

The Schiff base BSOPD was synthesized following the method described by Kismat *et al.,*[16] with slight modification. This was done by capping 1,2-diaminobenzene with salicylaldehyde in a 1:2 molar ratio in ethanol in the presence of 0.1 M NaOH (Scheme 1). The solution of the reactants was refluxed for 3 h at 55-65°C. Complexes of Nd(III) and Sm(III) were synthesized by refluxing solutions of the respectively salts with the Schiff base ligand in either 1:1 or 1:2 ratios in absolute ethanol for 6 h at 70°C. The solutions were concentrated to about 15.0 cm^3 and left in the refrigerator for precipitates to form. The precipitates were filtered, washed with successive portions of hot ethanol followed by diethyl ether. The complexes were left to dry in a desiccator over anhydrous $CaCl_2$[17]. The electronic transitions of complexes were recorded on a Jenway 3510 UV/visible spectrophotometer. The C, H and N elemental analysis was carried out using LECO-CHN-(#@ analyser. IR of the ligand and the complexes were recorded on Shimadzu FTIR MB 3000 infrared spectrophotometer using KBr pellet. The molar conductivity of the compounds with 10^{-1} M dimethylformamide (DMF) was done using LF-90; WTW conductivity meter. The results are presented in Table 1.

Antimicrobial Activity of BSOPD, Nd-BSOPD and Sm-BSOPD

Collection of Test Organisms: Clinical isolates for antimicrobial studies were obtained from microbial stock collection unit of the Postgraduate Laboratory Department of Microbiology, University of Uyo. The organisms were *Staphylococcus aureus*, *Bacillus subtilis, Escherichia coli, Pseudomonas aeruginosa, Candida albicans* and *Aspergillus niger*. These isolates were sub-cultured to obtain pure and fresh isolates that were used for this work. Few biochemical tests were carried out to confirm these isolates before being used.



2,2'-((1E,1'E)-(1,2- phenylenebis(azanylylidene))
bis(methanylylidene))diphenol

Scheme 1: Synthetic route for Schiff base ligand Bis(salicylaldehyde)orthophenylenediamine

Preparation of the test isolates before Inoculation

Pure cultures of these test organisms were inoculated into broth medium and incubated at 37°C for 24 h and 28°C for 72 h for bacterial and fungal isolates, respectively. All Gram+ve and fungal isolates were serially diluted to factor three using 10-fold dilution and Gram-ve isolates were serially diluted to factor 5 using 10-fold dilution. The least dilution for the isolates was compared with McFarland standard to march their turbidity with the standard[18].

Screening of the antimicrobial agents to check their activities on the isolates

The process was carried out using the well in agar diffusion technique. 0.1 mL of each diluted test organism was aseptically spread on the surface of the Muller-Hinton agar plates using a sterile hockey stick. The seeded plates were left for 30 min for isolates to diffuse into the medium. A sterile cork borer of 5 mm diameter was used to bore holes on the agar plates. Different concentrations of each sample were aseptically used to fill the agar wells such that each concentration was made to have contact with all the test organisms. All plates were incubated at 37°C for 24-48 h and 72 h -5 days for bacterial and fungal isolates, respectively. Antimicrobial activities of the samples against microbial pathogens were determined by measuring the zones of inhibition in mm. These tests were carried out in triplicate and the average values were recorded. Control experiments were set up alongside using the pure solvent.

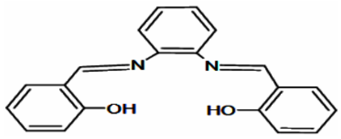
Minimum Inhibitory concentration (MIC)

The concentration of the sample that had the least activity for the zone of inhibition was further diluted to obtain different concentrations. 1 mL of these dilutions were added to 9 mL of sterile molten agar and poured into Petri dishes. The test organisms were inoculated by the streaking method. All the plates were incubated at 37°C for 24-48 hours. The least concentration that inhibits the growth of test organisms were recorded as minimum Inhibitory concentration (MIC) for that particular sample and the isolates.

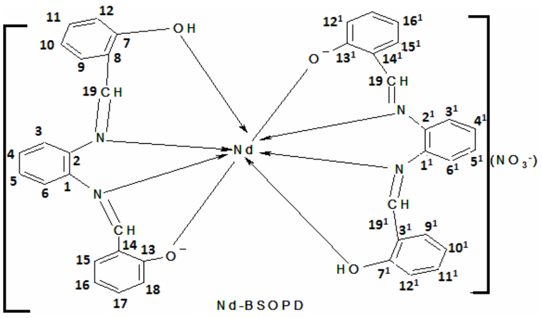
RESULTS AND DISCUSSION

Complexes of BSOPD with Nd(III) and Sm(III) ions were synthesized from ethanol. The physical properties of the complexes are presented in Table 1. The synthesized complex compounds are solids and are stable at room temperature. The melting points of the Schiff base and the metal complexes are listed in Table 1. These compounds are in powdery form. They are soluble in ethanol, dimethyl sulphoxide (DMSO) and dimethylformamide (DMF) but are insoluble in water. The elemental analysis of the Schiff base and its Ln (III) complexes listed in Table 1 are close to the calculated values based on the proposed molecular formulae. The molar conductivity of the DMF solutions (0.1 mol dm^{-3}) of the complexes ranged between 11.34-57.46 S cm^2 mol^{-1}. The low values obtained for Nd-BSOPD and Sm-BSOPD show that they are non-electrolyte complexes. This could be ascribed to the bonding of anions in these complexes rather than the ionic interaction with the lanthanide(III) cation in the course of complex formation. This unequivocally supports the non-electrolytic nature of these complexes,[19-21]. The absorption spectrum of the ligand BSOPD gave two prominent peaks at 262.00 and 272.00 nm assigned to π-π^* due to conjugated π bond of the phenyl ring and 333.00 nm assigned to n-π^* transition of conjugation between the lone pair of electrons of p-orbital of azomethine nitrogen and conjugated π bond of the benzene ring. This conforms with values given in the literature[22-24]. The electronic spectra of Sm-BSOPD showed a peak at 384 nm due to ligand to metal charge transfer (LMCT) but Nd-BSOPD showed three peaks at 318, 332 and 376 nm. It was observed that upon complexation, the absorption bands were shifted to longer wavelength (redshift) compared to the BSOPD ligand.

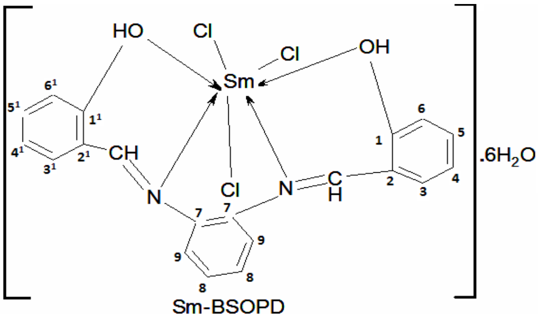
The characteristic IR bands of BSOPD and its Ln (III) complexes are listed in Table 2. The IR spectrum of BSOPD was compared with that of the Ln complexes to determine the coordination sites. The IR spectrum of the BSOPD ligand showed a strong band at 3456 cm^{-1} due to υ(O-H) of phenols. In the complexes, the υ(O-H) band became broad and occurred at 3342 cm^{-1} for Nd (III) ion while the bands for Sm (III) were within the range 3411-3404 cm^{-1}. This signifies the existence of a hydrogen bond as a result of lattice water. The presence of bands within 838-842 cm^{-1} also suggests the existence of coordinated water. The IR spectrum of BSOPD showed a strong band at 1616 attributed to υ(C=N) of azomethine. This band shifted to a higher wavenumber (difference of about 8-16 cm^{-1}) in all the complexes, signifying that the nitrogen of the azomethine is involved in chelation. This is also supported by bands appearing at 405-441 cm^{-1} in the metal complexes assigned to υ(Ln-N)[25-27]. The υ(C-O) is observed at 1282 cm^{-1} in BSOPD while in all the complexes, this band appeared at a lower wavenumber (difference of about 31-39 cm^{-1}). This suggests that BSOPD binds to Ln ions through phenolic oxygen. This is confirmed by the presence of bands around 622-649 cm^{-1} in all the complexes assigned to Ln-O[27]. Based on the elemental analysis, electronic spectra, and infrared spectral data the structures below are proposed for the ligand and complexes, respectively.



(a) 2,2'-((1E,1'E)-(1,2- phenylenebis(azanylylidene)) bis(methanylylidene))diphenol (BSOPD)



(b)



(C)

Fig. 1. (a) Structure of the ligand: 2,2'-((1E,1'E)-(1,2-phenylenebis(bis(azanylidene))bis(methanylylidene)) diphenol–BSOPD;(b) Proposed structure of Nd-BSOPD; (c) Proposed structure of Sm-BSOP

Table 1: Physical and Analytical data of BSOPD, Nd(III) and Sm(III) Complexes

Compound	F.W	Yield%	M:L ratio	m.p. Temperature°C	Colour	Molar conductivity Scm^{2}mol^{-1}	C% calc. (found)	H% calc. (found)	N% calc. (found)
BSOPD	316.36	77.34		158-160	Orange yellow	2.56	75.93 (75.97)	5.10 (5.06)	8.85 (8.91)
Nd-BSOPD $(C_{40}H_{30}N_5O_7Nd)$	836.95	60.00	1:2	150-152	Green	21.97	57.40 (57.48)	3.61 (4.31)	8.37 (8.84)
Sm-BSOPD $(C_{2}0H_{16}N_{2}O_{2}Cl_{3}Sm.6H_{2}O)$	681.16	62.00	1:1	140-142	Light brown	33.23	35.27 (35.95)	4.14 (4.12)	4.11 (4.41)

Table 2: IR Data of ligand and its Complexes

Compound	υ(O-H)	υ(C=N)	υ(C=C)	υ(C-O)	υ(C-H)	υ(Ln-N)	υ(Ln-O)	υ(Ln-Cl)
BSOPD	3456s	1616 s	1483 m	1282 s 1192 s	910 s	-	-	-
Nd-BSOPD	3342 br	1612 m	1458 m	1251 w 1153 w	910 w	426 m	649 w	-
Sm-BSOPD	3412 br	1626 m	1458 m	1248 m 1153 m	910 w	405 w	624 w	548 m

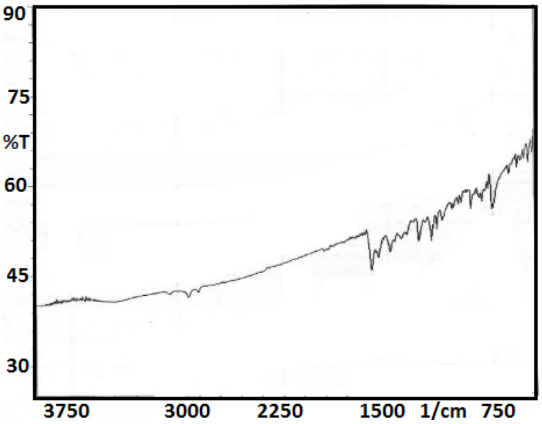


Fig. 2a. FTIR spectrum of the ligand (2,2'-((1E,1'E)-(1,2-phenylenebis(azanylylidene))bis(methanylylidene)) diphenol (Bis(salicylaldehyde)orthophenylenediamine)

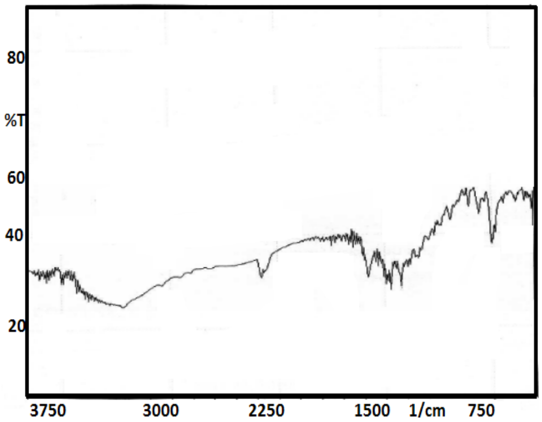


Fig. 2b. FTIR spectrum of Nd-Bis(salicylaldehyde) orthophenylenediamine

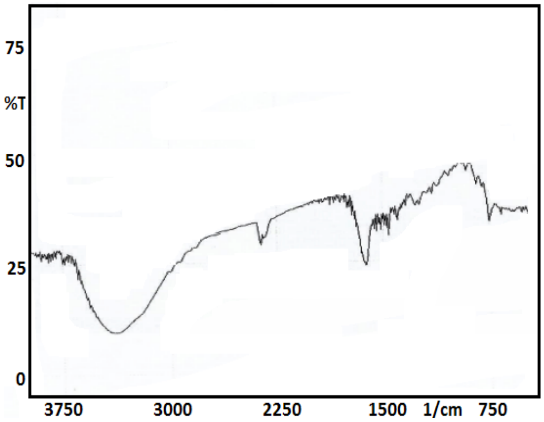
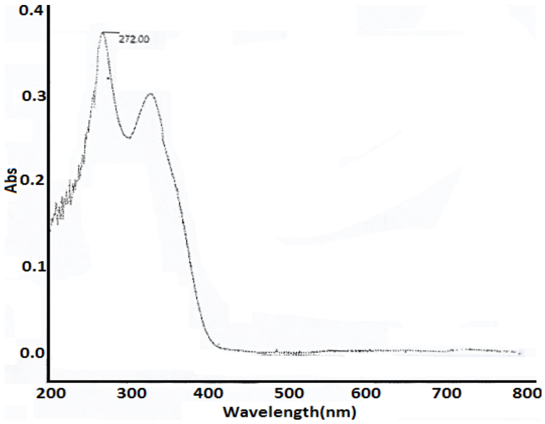
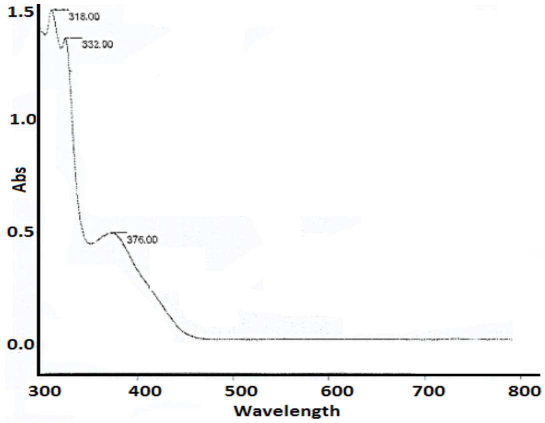


Fig. 2c. FTIR spectrum of Sm-Bis(salicylaldehyde) orthophenylenediamine

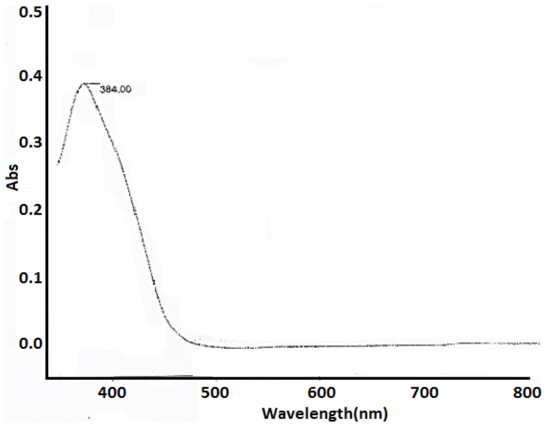
The pharmacological action of metal complexes depends greatly on the type of the metal ions and the donor order of the ligands because different ligands display diverse biological features. Numerous metal complexes are well-known to fast-track the drug action and the effectiveness of the organic therapeutic agents. The value of the various organic therapeutic agents can often be improved, upon coordination with an appropriate metal ion.



3a-(Bis(salicylaldehyde)orthophenylenediamine)



3b-Nd(Bis(salicylaldehyde)orthophenylenediamine)



3c-Sm(Bis(salicylaldehyde)orthophenylenediamine)

Fig. 3. UV-Visible spectra of BSOPD (a), Nd-BSOPD (b) and SmBSOPD (c)

The results presented in Table 3 specify that the ligand BSOPD and the complexes, Nd-BSOPD and Sm-BSOPD displayed good inhibitory effect against *Gram-positive* bacterium, *Bacillus subtilis* and *Gram-negative* bacterium *Escherichia coli*. The ligand and complexes also showed good antifungal activity against *Candida albicans* and *Aspergillus niger*. However, the ligand and the metal complexes had no effect on *Staphylococcus aureus* and *Pseudomonas aeruginosa* at all concentrations. The metal complexes showed better activity against *Bacillus subtilis* and *Aspergillus niger* than the ligand with activity increasing with an increase in concentration. The enhanced activity of the metal complexes compared to the ligand can be explained based on the chelation model. On complexation, the polarity of the metal ion is reduced due to the overlap of the ligand orbitals and partial sharing of the positive charge of the metal ion with donor groups. This increases the delocalization of π-electron over the chelate ring and augments the permeation of the complexes through the lipid layers of the microorganisms' membranes. It is advocated that factors such as solubility, conductivity, dipole moment, and cell permeability may also contribute to the increased activity of the complexes[28-34]. The antibacterial and antifungal effect against *E. coli* and *C. albicans*, respectively, were similar for both the ligand and the metal complexes, that is, the incorporation of the metal did not really improve the efficacy of the compounds against *E. coli* and *C. albicans*.

Table 3: Antimicrobial Screening Data for the Ligand and its Metal Complexes

Compound	Concentration	Diameter of inhibition zone(mm)			
	mg/mL	*B. subtilis*	*E. coli*	*C. albicans*	*A. niger*
BSOPD	20	12	11	11	10
	40	12	12	11	11
	60	15	14	14	16
	80	16	15	15	17
Nd-BSOPD	20	20	10	15	20
	40	21	12	15	15
	60	20	14	16	18
	80	22	15	15	17
Sm-BSOPD	20	18	13	12	18
	40	19	14	12	20
	60	21	15	14	21
	80	23	17	16	22
Streptomycin	20	5	2.5	-	-
Nystatin	10	-	-	31.3	62.5

Legend: (-) zones of inhition not included.

Table 4: Inhibition zones at 20mg/mL

Isolate	BSOPD	Nd-BSOPD	Sm-BSOPD
S. aureus	0	0	0
B. Subtilis	12	20	18
E. coli	11	10	13
P. aeruginosa	0	0	0
C. albicans	11	15	12
A.niger	10	20	18

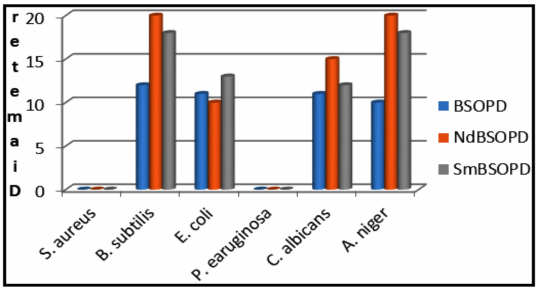


Fig. 4. Effect of ligands and their metal complexes toward isolates at Concentration of 20 mg/mL

Minimum inhibitory concentration

Minimum inhibitory concentrations were measured for the microoganisms that were affected by the compounds and the results obtained are presented in Table 5.

Table 5: Minimum inhibitory concentration of compounds (MIC, mg/mL)

Isolate	BSOPD	NdBSOPD	SmBSOPD
B. subtilis	10	10	10
E. coli	10	20	10
A. niger	20	5	10
C. albicans	20	10	20

The results of the MIC tests range from 5mg to 20 mg/mL. The lowest MIC was recorded for Nd-BSOPD against Aspergillu niger while BSOPD and Sm-BSOPD respectively recorded 20 mg/mL against *Candida albicans*. The results indicate that these compounds showed appreciable antimicrobial activity against these isolates.

CONCLUSION

Synthesis of 2, 2′-((1E, 1′ E)-(1, 2-Phenylenebis (Azanlylidene)bis(Methanylylidene)) Diphenol by the condensation of 1,2-diaminobenzene and salicylaldehyde in a 1:2 molar ratio was done successfully. The condensation product (Schiff base) was used to prepare Nd (III) and Sm(III) metal complexes. The prepared ligand and its metal complexes were characterised by physical and spectral techniques. IR spectral analysis indicates that the azomethine nitrogen and phenolic oxygen are linked to the central metal ion. On account of the CHN analysis and spectral information, the formulae $C_{20}H_{16}N_2O_2$, $[C_{40}H_{30}N_4O_4Nd]NO_3$ and $[C_{20}H_{16}N_2O_2Cl_3Sm].6H_2O$ are suggested for the Schiff base and metal complexes, respectively. *In vitro* antimicrobial activity revealed that the synthesized ligand and its metal complexes have significant antibacterial and antifungal activities against tested microorganisms with activity increasing with an increase in concentration.

ACKNOLEDGEMENT

The authors are thankful for the Heads of Departments of Pure and Industrial Chemistry, Faculty of Physical Sciences University of Nigeria, Nsukka and Akwa Ibom State University Ikot Akpaden, for laboratory facilities and to Mrs Udosen of the Department of Microbiology, University of Uyo for assisting with antimicrobial studies.

Conflict of interest

The authors declare that there is no conflict of interests regarding the publication of this paper.

REFERENCES

1. Alexander, V. *Chemical Reviews*., **1995**, *95* (2), 273-342.
2. Kulkarni, A.; Patil, S. A.; Badami, P. S. *European Journal of Medicinal Chemistry*. **2009**, *44*(7), 2904-2912.
3. Wanninger, S.; Lorenz, V.; Subhan, A.; Edelmann, F. T. *Chemical Society Reviews*., **2015**, *44*(15), 4986-5002.
4. Mohanan, K.; Kumari, B. S.; Rijulal, G. *Journal of Rare Earths*., **2008**, *26*(1), 16-21.
5. Kostova, I. *Current Medicinal Chemistry-Anti-Cancer Agents*., **2005**, *5*(6), 591-602.
6. Cundari, T. R.; Saunders, L. C. *Journal of Chemical Information and Computer Sciences*., **1998**, *38*(3), 523-528.
7. Fricker, S. P. *Chemical Society Reviews.,* **2006**, *35*(6), 524-533
8. Matsada, T.; Yamanaka, C.; Ikeya, M. *Applied Radiation and Isotopes*., **2005**, *62*(2), 353-357
9. Coelho, J.; Hussain, N. S.; Gomez, P. S.; Garcia, M. P.; Lopes, M. A.; Fernandes, M. H.; Santos, J. D. *Current Trends on Glass and Ceramic Materials*., **2012**, 87-115.
10. Grazul, M.; Budzisz, E. *Coordination Chemistry Reviews*., **2009**, *253*(21-22), 2588-2598.
11. Kostova, I.; Trendafilova, N.; Momekov, G. *Journal of Trace Elements in Medicine and Biology*., **2008**, *22*(2), 100-111.
12. Moksharagni, B.; Reddy, K. H. *International Journal of Pharmaceutical Sciences and Research*., **2017**, *8*(8), 3379-3384.
13. Hussain, A.; Chakravarty, A. R. *Journal of Chemical Sciences*., **2012**, *124*(6),1327-1342.
14. Venkatrmu, V.; Babu, P.; Jayasankar, C. K.; Troster, T.; Sievers, W.; Wortmann, G. *Optical Materials*., **2007**, *29*(11), 1429-1439.
15. Refat, M.S.; Al-Azab, F. M.; Al-Maydama, H. M. A.; Amin, R. R.; Jamil, Y. M. S. Spectro chima Acta Part A: *Molecular and Biomolecular Spectroscopy*., **2014**, *127*, 196-215.
16. Elachi, K. A.; Md.Hossain, S.; Md. Bitu, N. A.; Zahid, A. A. S. M.; Mohapatra, R. A.; Mannan, M. A.; Zakaria, C. M.; Md. Kudrat-E-Zahan. *Journal of Chemical, Biological and Physical Sciences*., **2019**, *9*(4), 201-218.
17. Uwanta, E.J; Ukoha, P.O. InterJournal *of Reseach and innovation in Applied Science*., **2019**, *4*, 67-69.
18. Ekong, U. S.; Mgbor, N. C.; Monece, A. N.; Obi, S. K. C. *Journals of Microbiology*., **2004**, *18*(1-2), 199-206.
19. Greary, W. *J. Coordination Chemistry Review*., **1971**, *7*, 81-122.
20. Gaye, M.; Tamboura, F. B.; Sall, A.S. *Bull. Chem.Soc. Ethiop*., **2003**, *17*, 27-34.
21. Naik, G.N. ; Bakale, R.P.; Pathan, A.H.; Ligade, S.G.; Desai, S.A.; Gudasi, K. B. *Journal of chemistry*., **2013**, article ID 810892,13 pages http//dx.doi.org/10.1155/2013/810892.
22. Taha, Z. A.; Ajlouni, A. M.; Al-Hassan, K.A.; Hajazi, A.K.; Faiq, A. B. *Spectrochimica Acta,* Part A., **2011**, *81*, 317-323.

23. Udinn, M. N.; Salam, M. A. *Research.*, **2014**, *1*, 777.
24. Cristovao, B.; Hnatejko, Z. *J. Molecular Structure.*, **2015**, *1088*, 50-55.
25. Liu, J.; Zhang, B.; Wu, B.; Hu, S. *Turk. J Chem.*, **2007**, *31*, 623-629.
26. Arora, K.; Sharma, M.; Sharma, K.P. *Coden Ecjhao E-journal of chemistry.*, **2009**, *6*(81), S201-S210.
27. El-Ansary, A. I.; Abdel-Kader. *Int. Jour. of inorganic chemistry.*, **2012**, Article ID90141513 pages doi:10. 1155/2012/901415
28. Chohan, Z.; Munawar, A.; Supuran, C. *Met. Based Drugs.,* **2001**, *8*, 137–143.
29. Iqbal, J.; Tirmizi, S.A.; Wattoo, F.H.; Imran, M. *Turk. J. Biol.*, **2006**, *30*, 1–4
30. Singh, V.P.; Katiyar, A.; Singh, S. *Biometals.*, **2008**, *21*(4), 491–501.
31. Ahamad, T.; Kumar,V.; Nishat, N. *Polymer International.,* **2006**, *55*(12), 1398–1406.
32. Patel, M. M.; Kapadia, M. A.; Patel, G. P.; Joshi, J. D. *Journal of Applied Polymer Science.*, **2007**, *106*(2), 1307–1317.
33. Neelakantan, M. A.; Marriappan, S. S.; Dharmaraja, J.; Jeyakumar, T.; Muthukumaran, K. *Spectrochimica Acta A: Molecular and Biomole.Spectroscopy.*, **2008**, *71*(2), 628–635
34. Bagihalli, G. B.; Patil, S. A.; Badami, P. S. *Journal of the Iranian Chemical Society.*, **2009**, *6*(2), 259–270