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Synthesis, characterization and antimicrobial activity studies of new Nickel(II) mixed ligand complexes of disubstituted dithiocarbamates with ethylsalicylaldiminate

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ABSTRACT

A new series of nickel(II) mixed ligand complexes of some disubstituted dithiocarbamates with ethylsalicylaldiminate have been synthesized and characterized. The complexes conform to the general formula, $[Ni(EtSal)(R_2dtc)]$ where EtSal = ethylsalicylaldiminate, $R_2 =$ dibenzyl (Bz_2), di-iso-butyl (i- Bu_2), di-n-butyl (n- Bu_2), ethylbutyl (EtBu), methylbutyl (MeBu), methylphenyl (MePh), cyclo-hexylmethyl (c-HxMe), and dtc = dithiocarbamate. Infrared spectra and elemental analyses reveal bidentate coordination of the ligands to nickel(II) ion in 1:1:1 mole ratio. Room temperature magnetic moments and solid reflectance spectra data indicate diamagnetic complexes with four-coordinate square planar geometry. The complexes show moderate and selective activity toward some of the test microorganisms.

Keywords: synthesis, characterization, disubstituted dithiocarbamates, ethylsalicylaldiminate, antimicrobial activity.

INTRODUCTION

Dithiocarbamates and their metal complexes have invited much research attention due to their diverse applications and interesting biological, structural, magnetic, electrochemical and thermal properties [1-7]. Metal complexes of dithiocarbamates with nitrogenous bases such as pyridine, picoline, 1,10-phenanthroline and 2,2'-bipyridine and with phosphine and its derivatives have been reported [8-16]. Literature reports on dithiocarbamate complexes containing Schiff bases are scarce [17]. Schiff bases are known to have biological activities such as antimicrobial [18-23], antifungal [18, 24], antitumour [25, 26] and as herbicides [27]. In view of the wide range of applications of dithiocarbamates [28] and various aspects of bioactivity of Schiff bases, we report in this article the synthesis and characterization of nickel(II) mixed ligand complexes of the type [Ni(EtSal)(R₂dtc)], where EtSal = ethylsalicylaldiminate (Schiff base) and R₂dtc =

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disubstituted dithiocarbamates. Antimicrobial activities of the complexes were investigated *in vitro*.

MATERIALS AND METHODS

Materials

All the chemicals were of analytical grade and commercially available from Sigma-Aldrich and British Drug Houses (BDH) Chemicals Limited. The reagents were used as received while the solvents were purified by standard methods [29]. The ethylsalicylaldimine ligand was prepared as the nickel(II) complex [30]. The various disubstituted dithiocarbamate ligands were formed by reacting carbon disulphide with the different disubstituted amines in ethanol and were used in that form without extracting from solution. Nutrient broth and Mueller Hinton agar were obtained respectively from Oxoid Limited, England and Lab M Limited, United Kingdom. The microorganisms used were supplied by the Department of Pharmaceutical Microbiology and Department of Botany and Microbiology, University of Ibadan, Nigeria.

Preparation of nickel(II) complexes, [Ni(EtSal)(R₂dtc)]

All the complexes were prepared by a typical reaction. A stirred mixture of stoichiometric amounts (2.2 mmol) of secondary amine, R_2NH and carbon disulphide, CS_2 in 10 mL ethanol was added dropwise to a stirring solution of 2.2 mmol bis(ethylsalicylaldiminato)nickel(II), Ni(EtSal)₂ in 20 mL warm ethanol. The reaction mixture was stirred for a further 1 hour. The precipitates formed were filtered under suction, washed with ethanol and dried *in vacuo* over silica gel.

Physical measurements

Microanalysis, infrared spectra and magnetic susceptibility measurements were performed at the Department of Chemistry, University of Zululand, KwaDelangezwa, South Africa. Elemental analyses were carried out on a Perkin Elmer 2400 Series II CHNSO Analyzer. The nickel content in the complexes was determined by complexometric titration. The melting points recorded were uncorrected. Infrared spectra were obtained on a Bruker FTIR Tensor 27 Spectrophotometer equipped with Silicon ATR, using pure samples of the compounds. Solid reflectance spectra were recorded on a Genesys 10 UV Scanning Spectrophotometer. Magnetic susceptibilities were measured at room temperature by Faraday method on a Sherwood Scientific Magnetic Susceptibility Balance calibrated Johnson Matthey with mercurv tetrathiocyanatocobaltate(II), $Hg[Co(SCN)_4]$. The data were corrected for diamagnetism by use of Pascal's constants.

Antimicrobial test

Antimicrobial test was performed on six bacteria (*Staphylococcus aureus, Bacillus subtilis, Escherichia coli, Pseudomonas aeruginosa, Proteus mirabilis* and *Klebsiella pneumoniae*) and four fungi (*Candida albicans, Candida glabrata, Candida tropicalis* and *Candida pseudotropicalis*). The media used were prepared by dissolving separately 2 g of the nutrient broth powder and 38 g of the Mueller Hinton agar powder in 250 mL and 1 L of deionized water, respectively. The two media were sterilized in an autoclave at 121 °C for 15 minutes and then stored overnight in a refrigerator after cooling. Cultures of the microorganisms were prepared in sterile nutrient broth and incubated for 24 hours at 37 °C for the bacteria and 27 °C for the fungi.

0.1 mL of each of the overnight cultures in sterile test tubes with caps were made up to 10 mL with 9.9 mL of sterile deionized water to give 1:100 or 10^{-2} dilution of the microorganisms.

The technique used for the study was agar-well diffusion. Solutions of concentration 10 mg mL¹ of the compounds were made in dimethylsulphoxide (DMSO). DMSO was also used as the negative control. The positive controls for bacteria and fungi were discs of commercial antibiotics manufactured by Abtek Biologicals Limited and fluconazole dissolved in DMSO. The discs were carefully placed on the innoculated media with the aid of sterile forceps. The plates innoculated with bacteria were incubated at 37 °C for 24 hours, and those innoculated with fungi were incubated at 27 °C for 72 hours. Afterwards, the zones of inhibition of microbial growth that appeared around the wells of the compounds were examined and the diameters measured and recorded in millimeters (mm).

				Elei	mental analys	sis, % Found	l (%
					Calcul	lated)	
Compound	Formula	Melting	Yield	Carbon	Undrogon	Nitrogan	Miekel
(Colour)	(Formula weight (g))	point (°C)	(%)	Carbon	Hydrogen	Nillogen	INICKEI
[Ni(EtSal)(Bz ₂ dtc)]	$C_{24}H_{24}N_2OS_2Ni$	146 149	72	60.52	5.44	6.73	12.45
(Green)	(479.29)	140–148	15	(60.14)	(5.06)	(5.85)	(12.25)
[Ni(EtSal)(i-Bu ₂ dtc)]	$C_{18}H_{28}N_2OS_2Ni$	129 120	74	52.67	6.52	7.26	14.38
(Dark green)	(411.26)	126-150	74	(52.57)	(6.88)	(6.81)	(14.28)
[Ni(EtSal)(n-Bu ₂ dtc)]	$C_{18}H_{28}N_2OS_2Ni$	154 156	16	52.25	6.21	7.60	13.94
(Dark green)	(411.26)	154-150	40	(52.57)	(6.88)	(6.81)	(14.28)
[Ni(EtSal)(EtBudtc)]	$C_{16}H_{24}N_2OS_2Ni$	06.08	80	50.22	6.36	7.55	15.16
(Dark green)	(383.20)	90-98	80	(50.15)	(6.33)	(7.31)	(15.32)
[Ni(EtSal)(MeBudtc)]	$C_{15}H_{22}N_2OS_2Ni$	103 104	84	48.63	6.09	7.74	16.18
(Dark green)	(369.58)	103–104	04	(48.74)	(6.01)	(7.58)	(15.89)
[Ni(EtSal)(MePhdtc)]	$C_{17}H_{18}N_2OS_2Ni$	150 160	00	53.10	4.98	7.21	15.28
(Green)	(389.17)	139-100	00	(52.46)	(4.67)	(7.20)	(15.09)
[Ni(EtSal)(c-	C H N OS Ni			51 67	6.73	7 35	15.07
HxMedtc)]	(205.21)	170-172	91	(51.67)	(6.13)	(7.00)	(14.86)
(Dark brown)	(393.21)			(31.00)	(0.13)	(7.09)	(14.00)

RESULTS AND DISCUSSION

Stoichiometry and physical characteristics

Analytical data and physical characteristics of the nickel(II) mixed ligand complexes of the various disubstituted dithiocarbamates with ethylsalicylaldiminate are listed in Table 1. The compounds were obtained in high yields (73 \square 91%) except [Ni(EtSal)(*n*-Bu₂dtc)] which gave a yield of 46%. Apart from [Ni(EtSal)(EtBudtc)] with a melting point of 96 \square 98 °C, all the complexes melted above 100 °C. The observed percentages for elemental composition are satisfactory and reveal 1:1:1 nickel to ligands stoichiometry which corresponds to the molecular formulae proposed for the compounds. Aside from [Ni(EtSal)(*c*-HxMedtc)] which is dark brown, the nickel(II) complexes are either green or dark green in colour. The compounds are generally insoluble in water, slightly soluble in ethanol and methanol, fairly soluble in dichloromethane and DMSO, and soluble in acetone, DMF and THF.

The general equation for the preparation of the nickel(II) dithiocarbamate-ethylsalicylaldiminate complexes, where $(R_2dtc) = (R_2NCS_2)$ is,

 $[Ni(EtSal)_2] + R_2NH + CS_2 \longrightarrow [Ni(EtSal)(R_2dtc)] + EtSalH$

Infrared spectra

The relevant bands in the infrared spectra of the mixed ligand complexes are listed in Table 2. The dithiocarbamate ligands show two diagnostic bands in the infrared spectra. First is the band due to $\nu C^{\dots}N$ thioureide vibration which is observed in the region 1494–1538 cm⁻¹. The high vibrational frequencies of this band in the mixed ligand complexes indicate a partial double bond character of the C $\Box N$ group arising from mesomeric drift of electrons from the dithiocarbamate moiety towards the nickel(II) ion center [11, 13] and the contribution of resonance structure (c) (Figure 1) of the thioureide form in accordance with [31]. The second single band which appears in the region 964–973 cm⁻¹ corresponds to $\nu C^{\dots}S$ vibration and indicates bidentate coordination mode of the dithiocarbamate moiety through the two sulphur donor-atoms [3].

The bands which appear in the regions $1596 \square 1607 \text{ cm}^{-1}$ and $1196 \square 1219 \text{ cm}^{-1}$ are assigned respectively to C=N and C \square O stretching frequencies in the ethylsalicylaldiminate ligand. The positions of the two bands indicate a bidentate coordination of the ethylsalicylaldiminate moiety to nickel(II) ion through the azomethine nitrogen [17] and phenolic oxygen [17, 32-34].

The nickel(II) dithiocarbamate-ethylsalicylaldiminate mixed ligand complexes show three diagnostic metal-ligand absorption bands in their infrared spectra. The bands which appear in the regions $378 \square 384 \text{ cm}^{-1}$, $461 \square 467 \text{ cm}^{-1}$ and $586 \square 596 \text{ cm}^{-1}$ are attributed to $\nu \text{Ni} \square \text{S}$, $\nu \text{Ni} \square \text{O}$ and $\nu \text{Ni} \square \text{N}$ vibrational modes, respectively and provide evidence of formation of the nickel(II) mixed chelates.

Compound	ν (C=N)	ν(C <u>···</u> N)	$\nu(C\Box O)$	$\nu(C^{\underline{\dots}}S)$	$\nu(Ni \Box N)$	$\nu(Ni\Box O)$	$\nu(Ni \square S)$
[Ni(EtSal)(Bz ₂ dtc)]	1605m	1494vs	1219vs	973s	586s	466vs	379vs
[Ni(EtSal)(<i>i</i> -Bu ₂ dtc)]	1606s	1499vs	1204s	972m	596m	467s	382vs
[Ni(EtSal)(n-Bu ₂ dtc)]	1606s	1507vs	1216s	967m	596m	466s	383vs
[Ni(EtSal)(EtBudtc)]	1606s	1515vs	1196m	964m	596m	467s	378vs
[Ni(EtSal)(MeBudtc)]	1607m	1516vs	1209vs	966s	596s	467m	378vs
[Ni(EtSal)(MePhdtc)]	1596m	1538m	1213s	966s	595s	466vs	384vs
[Ni(EtSal)(c-HxMedtc)]	1603vs	1512vs	1207vs	970s	592s	461vs	378vs

 Table 2. Infrared spectra data of the nickel(II) complexes

 $vs = very \ strong; \ s = strong; \ m = medium$



Figure 1. Resonance forms of dithiocarbamate complexes

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Electronic spectra and magnetic moments

The solid reflectance spectra data for the nickel(II) mixed ligand complexes are given in Table 3. The bands in the regions 40.16–40.98 kK and 30.12–30.49 kK are assigned respectively to intraligand $\pi \rightarrow \pi^*$ electronic transition in the N···C···S and S···C···S chromophores of the dithiocarbamate moiety [35-37]. The band in the region 36.50–37.31 kK is assigned to $\pi \rightarrow \pi^*$ transition in the ethylsalicylaldiminate moiety. Charge transfer bands are observed in the region 23.20–23.92 kK of the spectra. The two bands which appear in the regions 15.75–15.95 kK and 20.24–20.58 kK of the spectra of the compounds are consistent with square planar geometry [38] and are assigned to ${}^{1}A_{1g} \rightarrow {}^{1}A_{2g}$ ($b_{2g} \rightarrow b_{1g}$) and ${}^{1}A_{1g} \rightarrow {}^{1}B_{1g}$ ($a_{1g} \rightarrow b_{1g}$) electronic transitions, respectively. The magnetic moments further confirm that the nickel(II) complexes are diamagnetic with square planar geometry.

Compound	Band maxima, λ_{max} (kK)	Assignment	$\mu_{eff}(BM)$			
	15.95	$^{1}A_{1g} \rightarrow ^{1}A_{2g}(b_{2g} \rightarrow b_{1g})$				
[Ni(EtSal)(Bz.dtc)]	20.53	$^{1}A_{1g} \rightarrow ^{1}B_{1g}(a_{1g} \rightarrow b_{1g})$	Dia			
	23.42	Charge transfer	Dia			
	30.21, 37.31, 40.49	Intraligand transition				
	15.85	$^{1}A_{1g} \rightarrow ^{1}A_{2g}(b_{2g} \rightarrow b_{1g})$				
[Ni/EtSal)(; Dy. dta)]	20.37	$^{1}A_{1g} \rightarrow ^{1}B_{1g}(a_{1g} \rightarrow b_{1g})$	Dia			
	23.31	Charge transfer	Dia			
	30.49, 37.31, 40.98	Intraligand transition				
	15.87	$^{1}A_{1g} \rightarrow ^{1}A_{2g}(b_{2g} \rightarrow b_{1g})$				
[NI:/EtCol)/ Dr. dto)]	20.58	${}^{1}A_{1g} \rightarrow {}^{1}B_{1g}(a_{1g} \rightarrow b_{1g})$	Dia			
$[INI(EtSai)(n-Bu_2utc)]$	23.20	Charge transfer	Dia			
	30.49, 37.31, 40.98	Intraligand transition				
	15.75	$^{1}A_{1g} \rightarrow ^{1}A_{2g}(b_{2g} \rightarrow b_{1g})$				
[Ni/EtSal)/EtDudta)]	20.37	$^{1}A_{1g} \rightarrow ^{1}B_{1g} (a_{1g} \rightarrow b_{1g})$	Dia			
	23.47	Charge transfer	Dia			
	30.49, 36.90, 40.49	Intraligand transition				
	15.91	$^{1}A_{1g} \rightarrow ^{1}A_{2g}(b_{2g} \rightarrow b_{1g})$				
[Ni/EtSal)(Mapudta)]	20.37	$^{1}A_{1g} \rightarrow ^{1}B_{1g}(a_{1g} \rightarrow b_{1g})$	Dia			
[NI(EtSai)(MeBudic)]	23.47	Charge transfer	Dia			
	30.49, 37.31, 40.98	Intraligand transition				
	15.95	$^{1}A_{1g} \rightarrow ^{1}A_{2g}(b_{2g} \rightarrow b_{1g})$				
[Ni/EtSal)(MaDhdta)]	20.24	$^{1}A_{1g} \rightarrow ^{1}B_{1g}(a_{1g} \rightarrow b_{1g})$	Dia			
[INI(EtSal)(MePhatc)]	23.92	Charge transfer	Dia			
	30.21, 36.50, 40.16	Intraligand transition				
	15.82	$^{1}A_{1g} \rightarrow ^{1}A_{2g} (b_{2g} \rightarrow b_{1g})$				
	20.28	$^{1}A_{1g} \rightarrow ^{1}B_{1g}(a_{1g} \rightarrow b_{1g})$	D:-			
[Ni(EtSai)(c-HxMedtc)]	23.42	Charge transfer	Dia			
	30.12, 36.90, 40.98	Intraligand transition				
$kK = kiloKayser (1 \ kK = 1000 \ cm^{-1})$						

Table 3.	Solid reflectance s	spectra data and	magnetic moments	of the nickel(II)	complexes
I able 5.	Sond reneeunce a	pectra aata and	mugnetie momento	or the mener(II)	complexes

			<i>a</i>				,			
			Gr	owth inhib	ition zone	in millime	eters (m	m)		
			Ba	acteria						
	<u>((</u>	<u> Bram</u>	((lram negati	ive)	(Fungi)
	posit	tive)	<u> </u>	<u></u>		/	<u>.</u>			
Compound	<i>S</i> .	В.	Е.	Р.	Р.	К.	С.	С.	С.	С.
compound	aur	subt	coli	mirab	aerug	рпеи	alb	glab	trop	psdt
[Ni(EtSal)(Bz ₂ dtc)]	-	-	-	16	-	13	16	-	13	12
[Ni(EtSal)(<i>i</i> -Bu ₂ dtc)]	-	12	-	15	-	10	12	-	14	16
[Ni(EtSal)(<i>n</i> -Bu ₂ dtc)]	-	-	20	15	-	11	-	-	13	13
[Ni(EtSal)(EtBudtc)]	-	-	-	12	-	18	-	-	12	15
[Ni(EtSal)(MeBudtc)]	-	-	-	11	-	-	16	-	14	15
[Ni(EtSal)(MePhdtc)]	-	-	-	16	-	-	-	-	-	15
[Ni(EtSal)(c-		12		14		11			14	11
HxMedtc)]	-	12	-	14	-	11	-	-	14	11
CHL 30	19	14								
CXC 5	-	-								
ERY 5	-	-								
COT 25	15	-	-	-	-	10				
GEN 10	14	13	20	-	09	-				
AUG 30	-	-	-	-	-	-				
AMX 25	-	-	-	-	-	-				
TET 10/30	10	-	10	-	-					
NIT 300			-	-		20				
NAL 30			14	-	15	20				
OFL 30			35	-	31	27				
FLU 30							-	-	-	-
DMSO	-	-	-	-	-	-	-	-	-	-

Table 4.	Antimicrobial	activity data o	of the compounds	(10 mg mL^{-1})) and controls
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Resistance (-); Staphylococcus aureus (S. aur); Bacillus subtilis (B. subt); Escherichia coli (E. coli); Proteus mirabilis (P. mirab); Pseudomonas aeruginosa (P. aerug); Klebsiella pneumoniae (K. pneu); Candida albicans (C. alb); Candida glabrata (C. glab); Candida tropicalis (C. trop); Candida pseudotropicalis (C. psdt); Chloramphenicol 30 μ g (CHL 30); Cloxacillin 5 μ g (CXC 5); Erythromycin 5 μ g (ERY 5); Cotrimoxazole 25 μ g (COT 25); Gentamycin 10 μ g (GEN 10); Augmentin 30 μ g (AUG 30); Amoxycillin 25 μ g (AMX 25); Tetracyclin 10 μ g for Gram positive bacteria and 30 μ g for Gram negative bacteria (TET 10/30); Notrofurantoin 300 μ g (NIT 300); Nalidixic acid 30 μ g (NAL 30); Ofloxacin 30 μ g (OFL 30); Fluconazole 30 μ g (FLU 30); Dimethylsulphoxide (DMSO)

Antimicrobial activity

Antimicrobial activity of the nickel(II) mixed ligand complexes was evaluated *in vitro* against Gram positive bacteria, *Staphylococcus aureus* and *Bacillus subtilis*, Gram negative bacteria, *Escherichia coli, Pseudomonas aeruginosa, Proteus mirabilis* and *Klebsiella pneumoniae*, and fungi, *Candida albicans, Candida glabrata, Candida tropicalis* and *Candida pseudotropicalis*. The results for the complexes and commercial antibiotics used as positive controls are listed in Table 4. All the complexes show selective activity against three microorganisms - *Proteus mirabilis, Candida tropicalis* and *Candida pseudotropicalis* and selective inactivity against four others, *Escherichia coli, Staphylococcus aureus, Pseudomonas aeruginosa* and *Candida glabrata*. Generally, the complexes show moderate antimicrobial activity [39] which is independent of the solvent used.

CONCLUSION

Nickel(II) mixed ligand complexes of various disubstituted dithiocarbamates with ethylsalicylaldiminate have been synthesized and characterized. The two ligand moieties exhibit a bidentate coordination mode in the complexes. Solid reflectance spectra and magnetic data indicate that the nickel(II) complexes are diamagnetic and square planar (Figure 2). The complexes show selective activity towards some of the test microorganisms.



Figure 2. Structure of the nickel(II) complexes $(\mathbf{R} = \mathbf{C}_2\mathbf{H}_5)$

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