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Preparation, Characterization and Antibacterial Activity of Some Metal ion Complexes

M. M. EL-AJAILY*, F. A. ABDLSEED and S. BEN-GWEIRIF#

*Chemistry Department, Science Faculty, Garyounis University, Benghazi-Libya.

#Botany Department, Science Faculty, Garyounis University, Benghazi-Libya.

melagaily@yahoo.com

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Abstract: The Schiff base complexes derived from Salicylaldehyde and *o*-phenylenediamine have been prepared and characterized using several physical techniques, in terms; elemental analysis, molar conductance measurements, thermogravimetric analysis, magnetic moment measurements, infrared, electronic and electron paramagnetic resonance spectra. The elemental analysis data exhibit the formation of 1:1[M: L] complexes. The molar conductance values reveal a non- electrolytic nature. The thermogravimetric analysis data of Cr (VI) complex show the presence of water molecules. The obtained magnetic moment values exhibit the existence of three unpaired electrons in the Cr (III) complex and a diamagnetic phenomenon for the other three complexes. The infrared spectral data display the coordination behavior of the Schiff base towards Cr (VI), Cr (III), Pb (II) and TiO (IV) ions. The electronic absorption spectra of the Schiff base and its complexes show $\pi \rightarrow \pi^*$ (phenyl ring), $n \rightarrow \pi^*$ (HC=N) and the expected geometrical structure for the prepared complexes. The electron paramagnetic resonance spectral data satisfy the presence of a paramagnetic phenomenon and support the expected geometrical structure of Cr (III) complex. The Schiff base and its new complexes were tested for antibacterial activity against gram positive bacteria; *Staphylococcus aureus* and gram negative bacteria; *Salmonella*, *Escherichia coli* including the resistance bacteria *Pseudomonas aeruginosa*.

Keywords: Schiff base, Salicylaldehyde, *o*-Phenylenediamine, Complexes

Introduction

Complexes of some divalent metal ions with Schiff base derived from Salicylaldehyde and tyrosine have been prepared and investigated by using different physical tools and an octahedral structure was proposed for all complexes¹. Boghaei² prepared and characterized non-symmetrical tetra dentate Schiff base complexes of Ni(II) and Cu(II) ions. Maihub *et al.*³ Prepared and investigated some Schiff base complexes of Co (II), Ni (II) and Cu (II) ions. It is found that the prepared complexes have square planar structures.

The present paper aims to prepare a Schiff base (derived from the reaction of salicylaldehyde and *o*-phenylenediamine) and its complexes with Cr (VI), Cr (III), Pb (II) and TiO (IV) ions and illustrate their geometrical structures by using different techniques and also to study their antibacterial activity on some pathogenic bacteria.

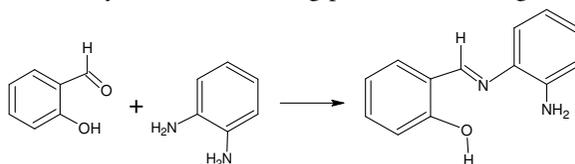
Experimental

Materials

All chemicals used in this work were reagent grade of BDH or Aldrich including CrO₃, CrCl₃.6H₂O, Pb(NO₃)₂, TiOSO₄, NH₄OH, C₂H₅OH, CHCl₃, DMF, NaOH, Salicylaldehyde, and *o*-phenylenediamine. Double distilled water were used throughout the experiment.

Preparation of Schiff base

The Schiff base under investigation was prepared by mixing an ethanolic solution (50 cm³) of 1.22 g; 0.01 mole of Salicylaldehyde with 1.08 gm; 0.01 mole of *o*-phenylenediamine in the same volume of ethanol. Few drops of 10 % NaOH were added to adjust pH and the obtained mixture then refluxed with stirring for two hours and the obtained precipitate was collected by filtration through Buchner funnel, recrystallized from ethanol, and dried at room temperature with 65% yield and its melting point is in the range of 191-197 °C.



Preparation of complexes

The complexes under investigation were prepared by mixing 50cm³ ethanolic solution of the Schiff base (2.12 gm; 0.01 mole) with the same amount of ethanolic solution of the metal salts (0.01 mole); CrO₃ (1.00 gm), CrCl₃.6H₂O (2.66 gm), Pb(NO₃)₂ (3.31 gm) and TiOSO₄ (1.59 gm). If the complexes did not separate, few drops of ammonium hydroxide were added to adjust the pH=8. The obtained mixtures were refluxed with stirring for extra four hours, and then filtered, collected and then washed several times with hot ethanol until the filtrate becomes clear. The complexes were dried in desiccators over anhydrous CaCl₂ under vacuum. The yield ranged from 60-75% and the melting points of all complexes are above 350 °C.

Bacterial culture

The strains of bacteria used were *Escherichia coli*, *Salmonella*, *Pseudomonas aeruginosa*, and *Staphylococcus aureus*. All strains were isolated from patients in Al-Jamahiriya hospital. The identity of all the strains was confirmed. A bacterial pension was prepared and added to the sterilized medium before solidification. The media with bacteria was poured into sterilized Petri dishes under aseptic condition. Different weights of Schiff base, Cr (VI),

Cr (III), Pb (II) and TiO (IV) Schiff bases complexes; (1mg, 5mg and 10mg) were placed on the surface of the culture and incubated at 37 °C for 24 h. After incubation the inhibition (mm) and the average of inhibition zones recorded.

Measurements

The prepared Schiff base and its complexes were subjected to (C, H, N and S) elemental analyses. The molar conductivity measurements were carried out in DMF solvent using conductivity meter model CMD650 digital. The magnetic moment measurements of the complexes were measured by using magnetic susceptibility balance Sherwood Scientific England. Infrared spectra were obtained by using KBr disk technique on IFS-25 DPUS/Ir spectrometer (Bruker) in the range of 4000-500 cm^{-1} . The electronic absorption spectra of the complexes were measured in chloroform solvent using a per kin- Elmer lambda 4 β spectrophotometer. The electron paramagnetic resonance spectrum of the Cr (III) complex was recorded by using EMX EPR spectrometer (Bruker) 1998Y at Al-Fateh University, Tripoli-Libya. The rest analyses were done at the Micro analytical center, Cairo- University.

Results and Discussion

Microanalysis

Table 1 comprises that the found data are in good agreement with those theoretical ones, and the obtained analytical analysis data indicate the formation of 1:1[M: L] ratio.

Molar conductivity

The conductance measurements of the prepared complexes were carried out in CHCl_3 solvent and the obtained values (Table 1) were taken as a good evidence for the existence of a non-electrolyte nature⁴.

Thermogravimetric analysis

The thermogravimetric analysis of TiO (IV) and CrO_3 complexes were performed to assist in predicting the molecular structures, the weight losses were measured from the ambient temperature up to 807 °C using a heating rate of 10 °C / min. The weight-losses of the complexes were calculated within the temperature range at which the water molecules were removed. The values were used in the calculation of the number of water molecules attached to the metal complexes. The weight-losses were calculated from the thermogravimetric curves and the calculated weight-losses were obtained from the suggested tentative formulae based on the elemental analysis data (Table 1). The weight loss of the $[\text{TiOLOH}]8\text{H}_2\text{O}$ complex occurring at 32% is attributed to the presence of 8 water molecules of hydration in the temperature range 0-520 °C. Whereas, for $[\text{CrO}_3\text{L}]12\text{H}_2\text{O}$ complex, the weight-loss at 40 % is due to the existence of 12 water molecules of hydration at temperature of 0-320 °C. The final products can be observed as metal oxides or carbonate at temperature above 520 °C.

Table 1. Some physical properties of the Schiff base and its complexes

Ligand/ complexes	M. Wt	C%		H%		N%		M.P °C	Λm^*
		calc	found	calc	found	calc	found		
$\text{C}_{13}\text{H}_{12}\text{N}_2\text{O(L)}$	212	73.58	73.70	5.66	5.20	13.20	13.00	191-197	-
$[\text{CrO}_3\text{L}].12\text{H}_2\text{O}$	528	29.54	29.84	6.81	6.22	5.30	5.23	>350	0.00
$[\text{CrL}.2\text{OH}.2\text{H}_2\text{O}].5\text{H}_2\text{O}$	406	38.42	38.45	5.17	4.70	6.89	6.87	>350	0.00
$[\text{PbL.OH}.2\text{H}_2\text{O}].6\text{H}_2\text{O}$	586	26.62	26.01	4.77	4.39	4.77	4.48	>350	0.00
$[\text{TiOL.OH}].8\text{H}_2\text{O}$	455	34.28	34.91	6.59	6.03	6.15	6.61	>350	0.00

Table 2. Thermogravimetric analysis data of Cr(III) complex

Complex	Water of hydration	Weight loss%	No. of water molecules	Temp °C	Metal oxide Weight loss %	Temp °C
[CrO ₃ L].12H ₂ O	(40.91)	40.00	12	0-320	(33.78) 33.33	>520

() = calculated value

Infrared spectra

The infrared band assignments of the Schiff base complexes of Cr (VI), Cr (III), Pb (II) and TiO (IV) ions (Table 3) exhibit a broad band in the range of 3163-3398 cm⁻¹ corresponding to the presence of water molecules.⁵ The bands in the range of 1608-1620 cm⁻¹ due the HC=N group and the change of these bands on complexation formation compared to its position in the free Schiff base (1614 cm⁻¹) suggesting the participation of azomethine group in coordination with the metal ions.⁶ New bands in the range of 447-609 and 536-756 cm⁻¹ are assigned to the ν (M-N) and ν (M-O) vibrations⁷. The appearance of these bands support the involvement of azomethine and hydroxyl groups via nitrogen and oxygen atoms in complexation. The spectra of [CrO₃L].12H₂O and [TiOL.OH].8H₂O complexes display two bands at 1020 and 1050 cm⁻¹, attributable to Cr =O and Ti=O bonds¹⁰. The band in the range of 3035-3350 cm⁻¹ indicates that the NH₂ group is not participate in coordination with the metal ions under investigation⁹.

Electronic spectra and magnetic moments

The electronic spectral data of the Schiff base and its complexes were recorded in chloroform and their assignments are listed in table 3. The spectral data of the Schiff base show three bands at 555 nm (18002 cm⁻¹), 550 nm (18182) and 333 nm (30030) due to $\pi \rightarrow \pi^*$ and $n \rightarrow \pi^*$ transitions¹⁰. The magnetic moment value of Cr (III) complex (6.12.BM) exhibits the existence of three unpaired electrons in the complex. The Cr (III) complex spectrum shows several bands (Table 3) due to ${}^4A_{2g}(F) \rightarrow {}^4T_{2g}(F)$, ${}^4A_{2g}(F) \rightarrow {}^4T_{1g}(F)$ and ${}^4A_{2g}(F) \rightarrow {}^4T_{1g}(P)$ transitions. An octahedral structure was suggested¹¹. The electronic spectral data of the [CrO₃L].12H₂O complex show three bands at 301nm (33222 cm⁻¹), 336 nm (29761cm⁻¹) and 373 nm (26809 cm⁻¹) attributed to charge transfer transition and an octahedral geometry was proposed for this complex¹². The electronic spectrum of [PbL.OH.2H₂O].6H₂O complex reveals two absorption bands at 303 nm (33057cm⁻¹) and 403 nm (24844 cm⁻¹) due to the presence of a charge transfer transition and an octahedral geometry was suggested for this complex¹³. For the [TiOL.OH].8H₂O complex, the electronic spectral data exhibit several bands (Table 3) corresponding to the existence of a charge transfer transition and an octahedral geometry was suggested¹⁴.

Electron paramagnetic resonance spectrum

The electron paramagnetic resonance spectrum of C(III) complex shows a g_{eff} value of 1.991. The small deviation of g_{eff} value than the ideal value (2.0023) results from the partial ionic character of the covalent bond between the Cr(III) ion and the Schiff base under investigation. This value supports the existence of an octahedral geometry¹² and confirmed the obtained data from the electronic spectra.

Table 3. Infrared spectral data of the Schiff base and its complexes

Schiff base/complexes	ν OH	ν NH ₂	ν C=N	ν C-O	ν M=O	ν M-O	ν M-N	ν OH (coord)	λ max nm, cm ⁻¹
C ₁₃ H ₁₂ N ₂ O (L)	3445	3083	1614	1276	-	-	-	-	555 (18002), 550 (18182), 333 (30030)
[CrO ₃ L]12H ₂ O	3398	3350	1612	1261	1020	667	540	2226	301(33221), 336 (29761), 373 (26809)
[CrL.2OH.H ₂ O]5H ₂ O	3323	3035	1620	1277	-	579	471	-	296 (33726), 318 (31397), 335 (29850), 375 (26631), 446 (22421)
[PbL.OH.2H ₂ O]6H ₂ O	3248	3059	1616	1254	-	536	447	-	303 (330033), 402 (24844)
[TiOL.OH]8H ₂ O	3163	3063	1608	1277	1050	756	609	-	307 (32573), 319 (31347), 332 (30120), 390 (25608), 427 (23419)

Antibacterial activity

Table 4 exhibits the zone of bacterial growth inhibition of the Schiff base which synthesized from Salicylaldehyde and 0-phenylenediamine and its complexes of Cr(VI), Cr(III), Pb(II) and TIO(IV) against the tested bacteria. The Schiff base (L) displays no effect against all bacteria tested. The Cr(VI) and Cr(III) complexes show no effect against all bacteria tested except *Pseudomonas aeruginosa* which was more sensitive to the all concentration used. The test results presented in table 4 show that Pb(II) complex had the strongest effect against both gram positive and gram negative bacteria in this study than all complexes used. However, the [TiOL.OH]8H₂O complex exhibits an antibacterial effect against all bacteria except *Staphylococcus aureus*. The results show the reduction of inhibition zone with the reduction of the compound weight placed on the bacterial culture. The study indicates that the Pb(II) solid complex has antibacterial activity against all bacteria used.

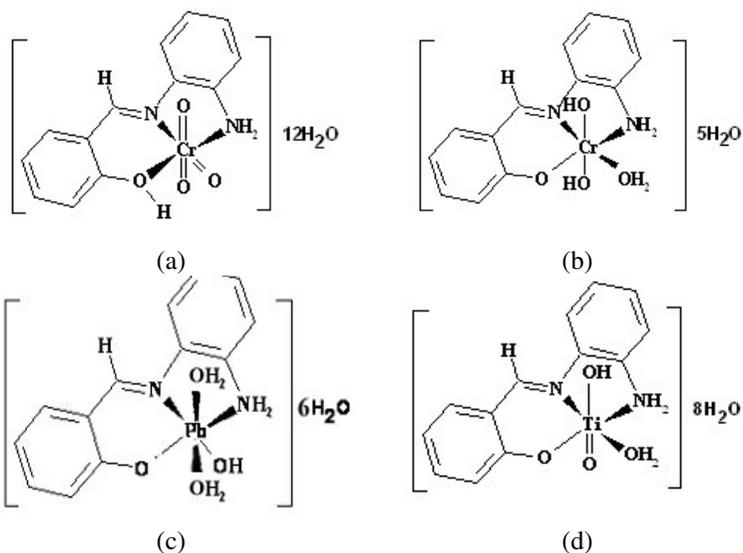
Table 4. The effect of Schiff base and its complexes on the bacterial growth

Compound/ Bacteria	<i>E. Coli</i> mg			<i>Salmonella</i> mg			<i>Pseudomonas</i> <i>Aeruginosa</i> , (mg			<i>Staphylococcus</i> <i>aureus</i> mg		
	1	2	3	1	2	3	1	2	3	1	2	3
C ₁₃ H ₁₂ N ₂ O (L)	-	-	-	-	-	-	-	-	-	-	-	-
[CrO ₃ L]12H ₂ O	-	-	-	-	-	-	-	+++	+++	-	-	-
[CrL.2OH.H ₂ O]H ₂ O	-	-	-	-	-	-	++	+++	+++	-	-	-
[PbL.OH.2H ₂ O]H ₂ O	-	+++	+++	-	+++	+++	++	+++	+++	+	+++	+++
[TiOL.OH]8H ₂ O	-	+	+	++	+++	+++	+	+++	+++	-	-	-

High active=+++ (inhibition zone >12mm); Moderately active = ++ (inhibition zone >9-12mm)
Slightly active =+ (inhibition zone >6-9mm); Inactive =- (inhibition zone<6mm)

Conclusions

From the previous chemical analyses, the following geometrical structures (a-d) were suggested.



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