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Minu G. Bhowon ^a & D. Nedhool ^a

^a Chemistry Department, Faculty of Science, University of Mauritius, Reduit, Mauritius Published online: 14 Apr 2008.

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SYNTHESIS AND CHARACTERIZATION OF SOME RUTHENIUM(II) SCHIFF BASE COMPLEXES

Minu G. Bhowon* and D. Nedhool Chemistry Department, Faculty of Science University of Mauritius Reduit, Mauritius

ABSTRACT

A number of ruthenium complexes, $RuL_2(H_2O)_2Cl_2$ (where L = monodentate Schiff bases derived from 3- and 4-anisaldehyde, 2-nitrobenzaldehyde, 4-chlorobenzaldehyde with 4toluidine and 4-aminobenzoic acid) and $RuL'(H_2O)(PPh_3)Cl_2$ (where L' = bidentate Schiff bases derived from 2-nitrobenzaldehyde or 3-hydroxybenzaldehyde with 3,4diaminotoluene), have been prepared. These complexes were characterized by elemental analyses, IR, UV-Vis, 'H NMR, conductance measurements and magnetic moments.

INTRODUCTION

A lot of research work is currently being undertaken in the field of synthesis of metal chelate complexes. These compounds continue to offer a wide scope for research work due to their preparative accessibility, diversity and structural variability¹.



Fig. 1. Structures of Schiff Base Ligands (1)-(8)

Schiff base complexes of transition metal ions of the first row have been widely studied². However, little has been reported on ruthenium(II) Schiff base complexes³. In the past few years, various authors have described the synthesis of novel, neutral, chiral Ru(II) Schiff base complexes derived from various aldehydes and amines^{4,5}. Recently, apart from their catalytic behaviour, the reversible binding of O₂ and CO to ruthenium(III) Schiff base complexes was also studied⁶⁻⁸.

Most of the papers reported in the literature on the preparation of ruthenium Schiff base complexes involved the use of $RuCl_2(PPh_3)_3$ as starting material^{3,9,10}. There are very few ruthenium Schiff base complexes which were prepared from $RuCl_3.xH_2O^{11,12}$. Our desire to synthesize ruthenium compounds containing Schiff base ligands has led us to the

preparation of these compounds by a route involving $RuCl_3.3H_2O$ in the presence of triphenylphosphine. The structures of the Schiff bases (1)-(8) used are shown in Fig. 1.

RESULTS AND DISCUSSION

The Schiff base ligands (1)-(8) were prepared by condensation of 3- or 4-anisaldehyde, 4chlorobenzaldehyde, 2-nitrobenzaldehyde, 3-hydroxybenzaldehyde with 4-aminobenzoic acid, 4-toluidine or 3,4-diaminotoluene in ethanol as white, yellow or brown solids. These ligands were characterized by elemental analyses, ¹H NMR and IR techniques with the data given in Table I. The imine structure of the Schiff bases is clearly indicated by intense IR bands near 1630-1590 cm⁻¹ and the presence of a NMR peak at δ 8.2-8.6 ppm confirmed the imine group in the Schiff bases. Green, brown or black microcrystals of ruthenium Schiff base compounds were made from RuCl₃.3H₂O and the Schiff bases (1)-(8) in the presence of triphenylphosphine. The stoichiometric compositions of the resulting mononuclear ruthenium(II) Schiff base complexes (1a)-(8a) are consistent with the microanalyses of the complexes (Table II). Millimolar solutions of the complexes in methanol are non-electrolytic in nature. Equation 1 represents the preparation of (1a)-(3a) and (7a)-(8a) while equations 2 and 3 represents the preparation of (4a), (5a) and (6a), respectively.

 $2RuCl_{3}.3H_{2}O + 4Schiff base + PPh_{3} \rightarrow 2[RuCl_{2}(Schiff base)_{2}(H_{2}O)_{2}] + OPPh_{3} + 2HCl + H_{2}O \qquad (1)$

 $2RuCl_{3}3H_{2}O + 2Schiff base + 3PPh_{3} \rightarrow 2[RuCl_{2}(Schiff base)PPh_{3}(H_{2}O)_{2}] + OPPh_{3} + 2HCl + H_{2}O$ $2RuCl_{3}3H_{2}O + 2Schiff base + 3PPh_{3} \rightarrow 2[RuCl_{2}(Schiff base)(PPh_{3})H_{2}O] + OPPh_{3} + 2RuCl_{3}(Schiff base)(PPh_{3})H_{2}O$

 $f_{2}O + 2SCHIII base + SFFH_3 \rightarrow 2[KuCl_2(SCHIII base)(FFH_3)f_2O] + OFFH_3 + 2[KuCl_2(SCHIII base)(FFH_3)f_2O] + 0[KuCl_2(SCHIII base)(FFH_3)f_2O] + 0[KuCl_3(FFH_3)f_2O] + 0[KuCl_3(FFH_3$

 $2HCl + 3H_2O$ (3)

10.8 (s, 1H), 8.27 (s, 1H), 6.5-8 (m, 8H), 3.7 (s, 3H) 8.9 (s, 2H), 7.2 - 7.85 (m, 11H). 2.5 (s, 3H) 9.5 (s, 2H), 7.8 (s, 2H), 6.2 - 7.5 (m, 11H), 2.3 (s, 3H) 8.45 (s, 1H), 7.1 - 7.8 (m. 8H), 2.4 (s, 3H) 8.4 (s, 1H), 6.8 - 7.8 (m, 8H), 3.9 (s, 3H), 2.4 (s, 3H) H NMR^b (δ, ppm) 12.6 (s, 1H), 8.39 (s, 1H), 6.8 -7.9 (m, 8H), 3.9 (s, 3H) 12.6 (s, 1H), 8.39 (s, 1H), 6.6 - 7.8 (m, 8H) IR data (cm⁻ v(C=N) v(C=0) 1631 m 1673 s 1619 m 1592 s 1681 s 1596 s 1677 s 1704 s 1589 s 1592 s 1681 s 1600 s 6.0 (6.1) (10.4)14.2 (14.4) 5.4 (5.4) 8.3 (8.4) 5.1 (5.4) 5.5 (5.4) (6.2) 10.3 **Elemental Analysis** Found (calc) (%) H N 6.3 4.1 (4.1) 5.1 (5.1) 6.8 (6.7) 5.1 (5.1) 3.6 (3.7) 5.4 (5.4) 5.3 (5.2) 3.6 (3.8) 80.1 (80.0) 70.0 (70.5) 70.1 (70.5) 62.0 (62.2) 64.4 (64.9) 76.1 (76.3) 72.8 (73.2) 64.3 (64.7) C GY = Greenish yellow, Y = Yellow, B = Brown, W = White M.P (°C) Colour² 62 S0 ۲ 168 ۲ 249 Y ۲ 108 ₹ 26 60 B 8 N ₹ <u>1</u>22 Yield (%) 75 88 78 35 96 61 9 82 C₂₁H₁₆N₄O₄ (<u>388</u>) p-BAB C₁₄H₁₀NO₂CI (259.5) CI4HI0N2O4 (270) m-BDA C₂₁H₁₈N₂O₂ (330) C₁₅H₁₃NO₃ (255) p-AAB C₁₅H₁₃NO₃ (255) p-ATA C₁₅H₁₅NO (225) m-AAB p-BTA C₁₄H₁₂NCI (229.5) Schiff bases (Formula o-BAB weight) Ξ 3 Ð E $\overline{\mathbb{C}}$ 3 € ⊛

Table I. Melting Points, Analytical, IR and ¹H NMR Data for the Schiff Bases

Table II. Melting Points, Elemental Analysis, IR and ¹H NMR, UV-Vis Data for the Ruthenium Schiff Base Complexes

								-
	Complexes	Yield	M.P (°C)	Elemental Analysis	LK data (cm ')	H NMR ² (0, ppm)	.siv - VU	_
	(Formula weight)	(%)	Colour"	Found (calc) (%)			λmax = nm	_
				C H N CI	v(C=N)		(E, M ⁻¹ cm ⁻¹)	_
					v(C=0)			
(12)	Ru(p-AAB) ₂ Cl ₂ (H ₂ O) ₂	40	176 (d)	49.6 3.8 3.8 10.8	1600 s	8.5 (d, 2H), 6.8 - 8.2 (m,	513 (3464), 346	_
	C30H30N2O8CI2Ru (718)		0	(50.1) (4.2) (3.9) (9.8)	1700 s	16H), 3.6 (s. 6H)	(44287), 308 (44551)	
(28)	Ru(p-ATA) ₂ Cl ₂ (H ₂ O) ₂	45	(p) 88 I	54.2 4.7 4.0 10.0		8.1 (d, 2H), 6.8 - 7.9 (m,	507 (3414)	_
	C ₃₀ H ₃₄ N ₂ O ₄ Cl ₂ Ru		æ	(54.7) (5.1) (4.2) (10.7)	1592 s			_
	(860)					נו) כיב - כיב (ווס או) כיב - סיב (H)		
(32)	Ru(m-AAB),Cl,(H,O),	47	115	50.0 4.1 3.8 9.8	1608 5	10.08 (s. 2H), 8.27 (s. 2H)	514 (3212). 274	_
	C30H30N2O6Cl2Ru (718)		в	(50.1) (4.2) (3.9) (9.8)	1700 s	6.5 -8 (m, 16H), 3.7 (s, 3H)	(21087)	
(48)	Ru(o-BAB)PPh ₃ Cl ₂ (H ₂ O) ₂	40	195	51.2 3.9 3.2 10.2	1608 s	8 (s, 1H), 6.6- 7.6 (m, 23H)	511 (4548), 289	
	C ₃₂ H ₂₉ N ₂ O ₆ PCl ₂ Ru		BI	(51.8) (3.9) (3.7) (9.5)	1700 s		(54642), 267	
	(740)						(14/48)	-
(52)	Ru(o-BDA)PPh ₃ Cl ₂ (H ₂ O)	45	611	54.0 3.6 6.4 8.5	1612 m	7 - 8 (m, 28H)	724 (1353)	_
	C36H33N4O5PCI2Ru (804)		n	(53.7) (4.1) (6.9) (8.8)				
(62)	Ru(m-BDA)PPh ₃ Cl ₂ (H ₂ O)	45	>250	58.0 4.2 3.1 9.8	1592 m	10.1 (s, 1H), 9.5 (s, 1H), 8.3	870 (1900)	
	C36H35N2O3Cl2PRu (746)		U	(57.9) (4.6) (3.7) (9.5)		(s, 2H), 7-8 (m, 26H), 2.4 (s, 3H)		
(72)	Ru(p-BTA) ₂ Cl ₂ (H ₂ O) ₂	54	138	50.7 4.1 3.8 20.5	1581 m	6.8 - 7.8 (m, 18H), 2.2 (s,	500 (5642), 400	_
	C28H28N2O2CLRu (667)		в	(50.3) (4.2) (4.2) (21.2)		6H)	(4338)	
(84)	Ru(p-BAB) ₂ Cl ₂ (H ₂ O) ₂	50	>300	46.5 3.0 3.4 19.5	1604 s	9.8 (s, 2H), 7.9 (d, 2H), 6.9-	512 (376)	_
	C ₂₈ H ₂₄ N ₂ O ₆ Cl ₄ Ru (727)		B	(46.2) (3.3) (3.8) (19.5)	1696 s	7.8 (m. 16H)		
0 = 0	rreen R = Brown Bl = Black							_

^b Solvent is d_a -dmso , s = singlet, m = multiplet, d = doublet ^c Solvent is methanol

RUTHENIUM(II) SCHIFF BASE COMPLEXES

IR Spectra

Water is retained by the complexes in the solid state, as indicated by v(OH) bands at 3300-3400 cm⁻¹ along with the deformation and rocking modes of water at 1620 and 620 cm⁻¹ and also by the elemental analyses and has been reported previously by Kureshy et al⁵. The v(C=N) vibration is shifted to lower wave numbers indicating the co-ordination of the azomethine nitrogen with the metal. The intense bands present at 1573, 1319 and 2919 cm⁻¹ are attributed to asymmetric and symmetric vibrations of v(NO₂) and v(CH₃), respectively, thus confirming their presence in the complexes. The complexes where triphenylphosphine is found to be coordinated to the metal center show all the characteristic bands of PPh₃ (1480, 1420, 1084, 744, 694 and 520 cm⁻¹ [v(Ru-P)].

¹H NMR Spectra

¹H NMR spectra of complexes (1a)-(8a) were recorded in DMSO-d₆, and show signals in the 6.8-8.2 ppm range which are assigned to the triphenylphosphine group and the aromatic protons of the respective Schiff base ligands (Table II). The azomethine proton at 8.2-8.9 ppm in all Schiff base ligands shows a downfield shift in (1a) and (6a) and in the others an upfield shift to 8.0-7.8 ppm, inferring co-ordination through the azomethine nitrogen atom. In (5a) and (7a) the signal of the azomethine proton was masked by the multiplet peak of the triphenylphosphine protons in the 7-8 ppm region. The signals at 3.2-3.9 ppm and 2.2-2.5 ppm are due to methoxy and methyl protons, respectively. In the case of (6a), signals at 10.1 and 9.5 ppm indicate the presence of hydroxyl protons.

UV-VIS Spectra and Magnetic Moments

The UV-Vis spectra of the ruthenium(II) complexes were obtained in methanol. In the complexes (5a) and (6a) (CH₂Cl₂) a broad band at 870 nm and 724 nm ($\varepsilon = 1900$ and

1353) was observed. This band is assigned to ligand to metal charge transfer or d-d transitions of ruthenium(II), as was reported in similar complexes^{6.8}. The broad band near 370-400 nm may be assigned to metal (d) to ligand (π^*) charge transfer. The bands or shoulders below 300 nm are of the intraligand, π - π^* or n- π^* , type¹³⁻¹⁶. The magnetic moments of all complexes indicate them to be low-spin d⁶ complexes [Ru(II)].

The proposed structures of the ruthenium(II) Schiff base complexes (1a)-(8a) are shown in Fig. 2.

Catalytic Oxidation of Cyclohexene

The oxidation of cyclohexene was carried out using (2a) in the presence of hydrogen peroxide as co-oxidant. The reaction was performed in dichloromethane and the amounts of products were monitored by gas chromatography using octane as the internal standard. Complex (2a) in the presence of hydrogen peroxide was found to oxidise cyclohexene to cyclohexene oxide, cyclohexen-1-ol and cyclohexen-1-one. Table III gives the results obtained in the oxidation of cyclohexene using (2a) as catalyst and H_2O_2 as co-oxidant. In Table IV, the results are summarised for the catalytic oxidation of piperonyl alcohol at room temperature using (3a)-(5a), (7a) and (8a) in the presence of N-methylmorpholine N-oxide as co-oxidant and 10 mL of dichloromethane/THF (1:1) as solvent. Molecular sieves have been used to remove water. The aldehydes formed after 3 hours of reaction were quantified as their 2,4-dinitrophenylhydrazone derivatives.

EXPERIMENTAL

All reagents were used without further purification. 4-Aminobenzoic acid, 4-toluidine, 3,4-diaminotoluene, 3-hydroxybenzaldehyde, 3- and 4-anisaldehyde, 4-chlorobenzaldehyde, 2-nitrobenzaldehyde and triphenylphosphine were all purchased



R =	= COOH, R ¹ = OM	e (1a)
	_ 1	

 $R = CH_3, R^1 = OMe$ (2a)

 $R = CH_3, R^1 = CI$ (7a)

$$R = COOH, R^{1} = CI$$
 (8a)









Fig. 2. Structures of Ruthenium Schiff Base Complexes

Table III. Catalytic Oxidation of Cyclohexene at Room Temperature using (2a) as Catalyst and H2O2 as Co-oxidants (Catalyst: 2x10⁻² mmol)

	Cyclohexen-1-one	1.5 (1.2)	10.0 (8.1)	11.7 (9.5)	23.0 (18.6)
erª (Yield ^b , %)	Cyclohexen-1-one	1.6 (1.3)	16.5 (13.4)	21.7 (8.1)	71.0 (58.3)
Тиглоv	Cyclohexene Oxide	10.6 (8.6)	4.1 (3.4)	3.2 (2.7)	3.2 (2.7)
	Reaction Time (hrs)	3	20	24	48

^a Mole of product per mole of catalyst;

Solvent: dichloromethane (10 mL) ^b Yield hased on alkene

Table IV. Oxidation of Piperonyl Alcohol at Room Temperature using Ruthenium Complexes as Catalyst and NMO as Co-oxidant

Turnover ^b	51.7	64.3	51.6	41.9	50.9
Yield of piperonal [*] (%)	72.0	86.2	77.5	81.6	84.8
Reaction Time (hrs)	3	3	3	3	3
Catalyst	(3a)	(4a)	(5a)	(7a)	(8a)

· Yield based on piperonyl alcohol

^b mole of product per mole of catalyst NMO = N-methylmorpholine N-oxide from BDH Chemicals or Aldrich. RuCl₃.3H₂O was obtained from Aldrich. Melting points were determined on an electro-thermal digital melting point apparatus. Infrared spectra were recorded on a Mattson 1000 FT-IR spectrometer in the range 4000-400 cm⁻¹. UV-Vis spectra were recorded on a PU 8700 series spectrometer. Elemental analyses were performed at Imperial College of Science, Technology and Medicine, London (U.K.) and at the Australian National University, Canberra (Australia). ⁻¹H NMR spectra were recorded at 250 MHz on a Bruker Electrospin spectrometer. Chloride ions were determined quantitatively by gravimetric analysis.

Preparation of the Schiff Bases

The ligands p-AAB (1), p-ATA (2), m-AAB (3), o-BAB (4), p-BTA (7) and p-BAB (8) were prepared by stirring one equivalent of aldehyde (0.022 mmol) and one equivalent of amine (0.022 mmol) in 20 mL ethanol at room temperature for one hour. A yellow crystalline solid was obtained which was filtered and washed thoroughly with ethanol and dried. For the ligands o-BDA (5) and m-BDA (6), one equivalent of amine and two equivalents of aldehyde in 20 mL ethanol were stirred at room temperature (refluxed for m-BDA) for 3 hours. The yellow o-BDA was washed with ethanol and recrystallised from hexane and dried <u>in vacuo</u>. For m-BDA, the solution obtained from the reaction was evaporated to a small volume. Upon addition of ether, a brown compound was precipitated. The compound was filtered and recrystallised in methanol. A brown compound was obtained and dried in vacuo.

Synthesis of the Complexes (1a)-(8a)

 $RuCl_{3.3}H_{2}O$ (1.0 mmol), Schiff base (2.0 mmol) and PPh₃ (0.13 g, 0.50 mmol), mole ratio 1:2:0.5, were dissolved in ethanol (20 mL) and refluxed for nine hours under a

nitrogen atmosphere. The obtained solution was concentrated on a rotatory evaporator. Upon addition of ether, a precipitate was formed. It was filtered and recrystallised in 10 mL methanol/20 mL ether. The resulting compounds were redissolved in methanol. Addition of ether and further stirring yielded the desired microcrystalline compounds. The ruthenium complex (4a) with the Schiff base o-BAB was prepared by the above method with the molar ratio RuCl₁.3H₂O, Schiff base, PPh₃ 1:1:1.5.

The ruthenium complexes (5a) and (6a) with the Schiff bases o-BDA and m-BDA were prepared by the above method with the molar ratio $RuCl_3.3H_2O$, Schiff base, PPh₃ 1:1:1.5.

Catalytic Oxidation of Cyclohexene

The catalyst (1.5x10⁻² mmol) was dissolved in 30 mL of dichloromethane and then the substrate cyclohexene (0.15 mL, 1.83 mmol) was added. Octane was used as internal standard. Hydrogen peroxide was added in excess as a co-oxidant. The oxidation was monitored by gas chromatography. The column was maintained at 65 °C. The reaction mixture was stirred continuously at room temperature for 48 hours.

Catalytic Oxidation of Piperonyl Alcohol

To a solution of piperonyl alcohol (0.152 g, 1 mmol) in 10 mL of dichloromethane/THF (1:1) was added N-methylmorpholine N-oxide (0.351 g, 3 mmol) and the ruthenium Schiff base complex ($1-2x10^{-2}$ mmol). The solution was stirred for 3 hours at room temperature in the presence of 4Å molecular sieves (0.5 g). The mixture was filtered and the filtrate was evaporated to dryness and extracted with diethyl ether (2x25 mL). The combined ether extracts were filtered and evaporated to give the corresponding aldehyde which was then quantified as the 2,4-dinitrophenylhydrazone.

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