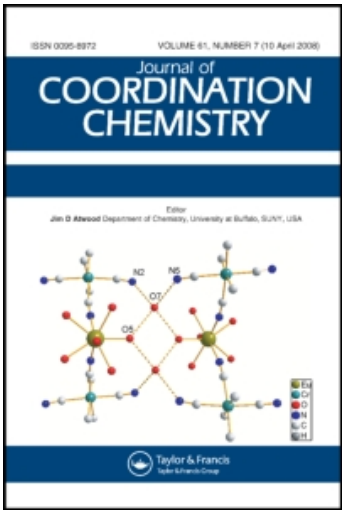

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Complexes of substituted dipyridylpyridazines with palladium(II) and platinum(II)

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Reactions of 3,6-*bis*(2′-pyridyl)pyridazine derivatives (*n*-dppn)¶ with $MX_2(PhCN)_2$ (M = Pd, Pt; X = Cl, Br) have been investigated. The new complexes *cis*-[$PdCl_2$(*n*-dppn)] (*n* = 5, 6, 8, 12), *cis*-[$PtCl_2$(*n*-dppn)]·H_2O (*n* = 5, 6), *cis*-[$PtCl_2$(8-dppn)] and *cis*-[$PtBr_2$(5-dppn)] have been characterized by elemental analyses, conductivity measurements, infrared, electronic and ^{1}H-NMR spectra.

Keywords: Pd(II) and Pt(II) complexes; Substituted dipyridylpyridazines derivatives

1. Introduction

Complexes of palladium(II) and platinum(II) with nitrogen-donor ligands are of great importance for their possible use in chemotherapy [1], catalysis [2], photoemission and redox reactions [3].

Moreover, pyridyl-containing ligands are under intense investigation due to their interesting, spectroscopic, photo-chemical and photo-physical properties as well as their use in building up supramolecules [4].

The tetranitrogenated 3,6-*bis*(2′-pyridyl)pyridazine (dppn) ligand, which is expected to be a better π-acceptor than 2,2′-bipyridine (bpy) and 1,10-phenanthroline (phen) [5, 6], has been reported to form complexes with Pd(II) and Pt(II) [7]. Such complexes have been used to synthesize bimetallic heteronuclear species [7d].

Although complexes of Ru(II) [8] and Rh(III) [5] with derivatives (*n*-dppn, figure 1) of the dppn ligand have been synthesized and characterized, no analogous complexes with Pd(II) and Pt(II) have been reported. The *n*-dppn ligands are expected to be even better donor than dppn itself due to the presence of the electron-releasing cyclic aliphatic groups [5, 8, 9].

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¶For the *n*-dppn ligands, *n* stands for the size of the cyclic aliphatic ring on positions 4 and 5 of the pyridazine ring, *n* = 5, 6, 8, and 12.

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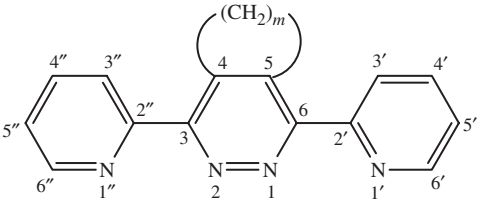


Figure 1. Structure of the ligands: 5-dppn, $m = 3$; 6-dppn, $m = 4$; 8-dppn, $m = 6$; 12-dppn, $m = 10$.

As a part of our investigation of the reactions of Pd(II) and Pt(II) with α-diimine ligands, we report here the isolation and characterization of mononuclear *cis*-dihalo complexes of Pd(II) and Pt(II) with the *n*-dppn ligands.

2. Experimental

2.1. *Materials*

All solvents used were AR grade. The metal salts and the starting materials for the preparation of the ligands were obtained from BDH, Aldrich, Fluka or Laborat. The substituted dipyridylpyridazines (*n*-dppn) were prepared according to the literature [9]. The starting benzonitrile complexes $[MX_2(PhCN)_2]$ (M = Pd, Pt; X = Cl, Br) were synthesized as reported earlier [10].

2.2. *Physical measurements*

Elemental analyses for the complexes were run by M-H-W Laboratories, Phoenix, Arizona, USA. Conductivity measurements were carried out on a Jenway 4010 digital conductivity meter at 25°C using 1.0×10^{-3} M solutions in *N*,*N*-dimethylformamide (DMF). The infrared absorption spectra (KBr and CsI pellets) were recorded on a Pye-Unicam SP3-300 spectrophotometer. Electronic absorption spectra were measured using a CARY 2390 spectrophotometer in CH_3CN. The ^{1}H-NMR spectra were carried out on a 400 MHz Varian VXR 400 spectrometer. Chemical shifts were referenced to tetramethysilan (TMS). The NMR spectra were obtained in deuterated dimethylsulfoxide (DMSO-d_6).

2.3. *Preparation of the complexes*

The complexes were prepared according to the following general procedures:

2.3.1. *cis*-[$PdCl_2$(*n*-dppn)] (*n* = 5, 6, 8, 12). A solution of $[PdCl_2(PhCN)_2]$ (0.5 mmol) in acetone (15 mL) was added dropwise to a stirred solution of the ligand (0.5 mmol) in acetone (30 mL). A precipitate formed immediately. The mixture was stirred at room temperature until the completion of the reaction which was monitored by the disappearance of the starting materials using thin layer chromatography (TLC) by

chloroform eluent. The product was filtered off, washed well with acetone and petroleum ether (20–40°C) and then dried under vacuum at room temperature.

2.3.2. *cis*-[PtX_2(*n*-dppn)] (*n* = 5, 6, 8; X = Cl and *n* = 5; X = Br). These complexes were prepared by reacting the *n*-dppn ligands with [$PtX_2(PhCN)_2$] (X = Cl, Br) as above except that chloroform was used as a solvent instead of acetone and the reaction mixture was refluxed instead of stirring at room temperature.

3. Results and discussion

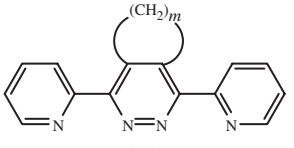
The tetranitrogenated dipyridylpyridazine derivatives *n*-dppn ($n = 5, 6, 8, 12$) were reacted with [$PdCl_2(PhCN)_2$] and [$PtX_2(PhCN)_2$] (X = Cl, Br) in acetone and chloroform, respectively, using 1 : 1 metal-to-ligand ratios. The results are shown in tables 1–4.

Analytical and physical data for the prepared complexes are presented in table 1. The solid compounds are all colored, stable in air and were isolated in moderate to good yields (33–60%). The complexes are insoluble in water while they are soluble to sparingly soluble in DMF and DMSO. All the complexes are non-electrolyte in DMF as shown in table 1. The slight conductance observed may be due to some dissociation or solvolysis in solution [11].

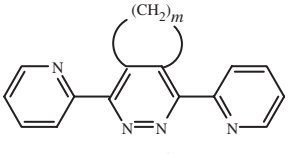
3.1. *IR spectroscopy*

Important infrared bands for the ligands and their complexes are listed in table 2. These bands were assigned with the aid of the reported data for the free *n*-dppn ligands and their complexes [5, 7b,f, 8, 9].

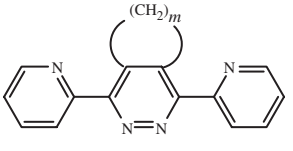
We reported earlier [5] that the free ligands may exist in three different conformations as shown below



cis-cis



trans-cis



trans-trans

However, the ^{1}H-NMR spectral measurements (table 4) indicate that the ligands exist in either the *cis–cis* or the *trans–trans* conformation and not only the former, as has been reported [5]. The latter conformation is even more favored since it has less lone-pair/lone-pair repulsion. Moreover, X-ray studies of the 3,6-*bis*(2′-pyridyl)pyridazine complexes show that the free nitrogens are *trans* while they adopt a *cis* conformation upon complexation [7a,c,f,g,h, 12]. Thus a detailed comparison cannot be made between free and coordinated ligands since they may have different conformations.

In the present study, the main bands that undergo change upon coordination are the pyridazine and pyridine bands at 1550–1585 and 990 cm^{-1}, respectively, which shift to higher frequencies and/or reduce in intensity (table 2). Complexes of the unsubstituted dppn ligands have been reported to show similar behavior [7b,d].

Table 1. Analytical and physical data for the complexes.

Complex	Anal. Calcd (found)% C	H	N	M.p. °C (Dec.)	Color	Yield (%)	Λ_M (DMF) ($ohm^{-1} cm^2 mol^{-1}$)
cis-[$PdCl_2$(5-dppn)]	45.2 (45.3)	3.1 (3.4)	12.4 (12.3)	297	Yellow	53	3.9
cis-[$PdCl_2$(6-dppn)]	45.4 (45.4)	3.5 (3.8)	12.0 (12.1)	298	Yellow	57	2.1
cis-[$PdCl_2$(8-dppn)]	48.7 (48.7)	4.1 (4.4)	11.4 (11.4)	306	Yellow	59	5.4
cis-[$PdCl_2$(12-dppn)]	52.4 (52.9)	5.1 (5.6)	10.1 (10.3)	202	Yellow	58	8.2
cis-[$PtCl_2$(5-dppn)]·H_2O	36.6 (36.9)	2.9 (2.9)	10.0 (9.9)	250	Green	38	11.0
cis-[$PtCl_2$(6-dppn)]·H_2O	37.8 (37.6)	3.2 (3.4)	9.8 (9.7)	280	Yellow	60	9.2
cis-[$PtCl_2$(8-dppn)]	41.2 (41.4)	3.5 (3.6)	9.6 (9.5)	260	Yellow	43	12.4
cis-[$PtBr_2$(5-dppn)]	32.5 (32.3)	2.2 (2.2)	8.9 (8.6)	334	Yellow	57	10.8

Table 2. Important IR frequencies for the ligands and their complexes (KBr and CsI pellets; cm^{-1}).

Compound	Ring breathing vibrations	Py ring vibrations	ν(M–X)[a]
5-dppn[b]	1580s, 1570m, 1460m, 1440m, 1370m	990m	
6-dppn[b]	1580s, 1560s, 1550m, 1470s, 1420m, 1380m	990m	
8-dppn[b]	1550m, 1480m, 1375m	990m	
12-dppn	1570s, 1525s, 1460m, 1430m, 1415m, 1380m	990m	
cis-[$PdCl_2$(5-dppn)]	1590m, 1530w, 1480m, 1440m, 1380m	1100m	355sh, 330m
cis-[$PdCl_2$(6-dppn)]	1560m, 1500m, 1470m, 1420m	1090m	390w, 320s
cis-[$PdCl_2$(8-dppn)]	1590m, 1500m, 1470m	1090m, 1030m	335sh, 320s
cis-[$PdCl_2$(12-dppn)]	1575m, 1520m, 1460m, 1360m	1100m	360sh, 330s
cis-[$PtCl_2$(5-dppn)]·H_2O[c]	1580m, 1490m, 1420w	1100m	340s, 330s (split)
cis-[$PtCl_2$(6-dppn)]·H_2O[c]	1590w, 1550m, 1460m, 1390m	1090m	355sh, 325s
cis-[$PtCl_2$(8-dppn)]	1586m, 1500m, 1470m, 1470m, 1320w	1100m	340s, 325s (split)
cis-[$PtBr_2$(5-dppn)]	1580m, 1530m, 1480m, 1470m, 1380m	1080m	270w, 260sh

[a]X = Cl, Br; [b]Ref. [5]; [c]ν(O–H): 3450 br.m.

The appearance of broad, medium absorption bands at 3450 cm^{-1} in the hydrated complexes supports the presence of lattice water [13a].

The presence of two ν(M–X) bands (table 2) suggests a *cis* configuration [2c, 13b, 14] for all these complexes as shown in figure 2.

3.2. *Electronic spectra*

The electronic spectra of the free *n*-dppn ($n = 5, 6, 8$) ligands have been reported [5, 8] to show two bands in the 217–287 nm range. Similarly, the 12–dppn ligand exhibits two bands at 267, and 217 nm.

The electronic spectra of the prepared complexes are characterized by the presence of strong ligand-centered (LC) bands along with some metal-ligand

Table 3. Electronic absorption spectra (in CH_3CN) for the ligands and complexes.

Compound	λ_{max} (nm)	$\varepsilon \times 10^{-3}$ ($L\,mol^{-1}\,cm^{-1}$)	Band assignments
5-dppn[a]	287	12.5	LC
	230sh	8.5	LC
6-dppn[a]	270	5.5	LC
	217	8.7	LC
8-dppn[a]	267sh	8.0	LC
	220	15.8	LC
12-dppn	267sh	5.0	LC
	217	10.4	LC
cis-[$PdCl_2$(5-dppn)]	309	17.5	LC
	430	2.5	MLCT
cis-[$PdCl_2$(6-dppn)]	280	39.3	LC
	406	8.9	MLCT
	462	5.4	MLCT
cis-[$PdCl_2$(8-dppn)]	280	11.0	LC
	304	6.9	LC/MLCT
	425	0.6	MLCT/d–d
	462sh		MLCT
cis-[$PdCl_2$(12-dppn)]	272sh	28.4	LC
	303	19.6	LC
	461	0.8	MLCT/d–d
cis-[$PtCl_2$(5-dppn)]$\cdot H_2O$	290	42.3	LC
	391	9.1	MLCT
	457	5.8	MLCT
cis-[$PtCl_2$(6-dppn)]$\cdot H_2O$	339	39.5	LC
	427	11.2	MLCT
cis-[$PtCl_2$(8-dppn)]	297	32.4	LC
	407	9.8	MLCT
	460	5.9	MLCT
cis-[$PtBr_2$(5-dppn)]	306	39.7	LC
	410	12.6	MLCT

[a] Ref. [5].

charge-transfer (MLCT) and ligand-field (d–d) absorption bands (table 3). The weak ligand-field (d–d) could not be observed for most of the complexes since they may be masked by the strong broad MLCT bands. The MLCT and d–d transitions are responsible for the characteristic color of d^8 metal ions [15, 16].

3.3. *^{1}H NMR*

The ^{1}H NMR spectral data of the *n*-dppn ligands and some of their complexes are presented in table 4. Problems with the solubility of other complexes prevented the measurement of their spectra. The peaks were assigned by comparison with ^{1}H-NMR data reported for the free *n*-dppn ligands and their complexes [5] as well as other related diimines [2c, 14, 17].

The ^{1}H NMR spectra for the *n*-dppn ($n = 5, 6, 8$) ligands (table 4) suggest four different types of protons for the pyridine hydrogens. Such results are consistent with the symmetrical *cis–cis* or *trans–trans* conformations and exclude the unsymmetrical *cis–trans* conformation. On the other hand, the ^{1}H NMR spectra for the complexes (table 4) show eight different peaks for eight different protons of the pyridine rings. The spectra also exhibit bands in the 1.16–3.60 ppm region, which are characteristic of aliphatic protons [5, 18].

Table 4. ^{1}H NMR characteristics of ligands and their complexes.[a]

Compound	NMR band shift: δ, ppm (J, Hz)
5-dppn[b]	8.78 (br.s, 2H), 8.49 (br.s, 2H), 8.05 (br.s, 2H), 3.42 (br.s, 4H), 2.08 (t, 2H, $J=6.8$)
6-dppn[b]	8.74 (d, 2H, $J=5.2$), 8.04 (t, 2H, $J=7.6$), 7.90 (d, 2H, $J=7.6$), 7.54 (q, 2H, $J=4.8$, 7.6), 2.89 (br.s, 4H), 1.73 (br.s, 4H)
8-dppn[c]	8.71 (m, 2H), 7.75 (m, 4H), 7.34 (m, 2H), 3.57 (t, 4H), 1.57 (2m, 8H)
cis-[$PdCl_2$(6-dppn)]	9.39 (d, 1H, $J=5.6$), 8.77 (d, 1H, $J=4.4$), 8.55 (d, 1H, $J=8.0$), 8.41 (t, 1H, $J=8.0$), 8.31 (t, 1H, $J=7.8$), 8.05 (d, 1H, $J=7.6$), 7.39 (t, 1H, $J=6.4$), 7.63 (t, 1H, $J=6.4$), 3.27 (br.s, 2H), 3.17 (br.s, 2H), 1.81 (m, 4H)
cis-[$PdCl_2$(8-dppn)]	9.41 (br.s, 1H), 8.78 (br.d, 1H), 8.43 (d, 1H, $J=6.8$), 8.37 (d, 1H, $J=6.4$), 8.10 (t, 1H, $J=7.6$), 8.06 (t, 1H, $J=7.2$), 7.94 (t, 1H, $J=5.2$), 7.63 (t, 1H, $J=5.2$), 3.30 (br.s, 2H), 3.11 (br.s, 2H), 2.00 (br.s, 2H), 1.90 (br.s, 2H), 1.60 (br.s, 2H), 1.45 (br.s, 2H)
cis-[$PtCl_2$(5-dppn)]$\cdot H_2O$	8.86 (d, 1H, $J=6.4$), 8.59 (br.d, 1H), 8.46 (t, 1H, $J=7.6$), 8.20 (t, 1H, $J=7.6$), 7.89 (2d, 1H, $J=8.4$), 7.74 (d, 1H, $J=7.6$), 7.62, (t, 1H, $J=6.4$), 7.47 (t, 1H, $J=8.0$), 3.60 (br.t, 2H), 3.52 (br.t, 2H), 2.30 (br.s, 2H)
cis-[$PtCl_2$(6-dppn)]$\cdot H_2O$	9.81 (d, 1H, $J=5.2$), 8.32 (dt, 1H, $J=4.4$, 17.6), 8.60 (d, 1H, $J=8.8$), 8.47 (t, 1H, $J=8.4$), 8.15 (m, 1H), 7.96 (t, 1H, $J=7.6$), 7.67 (t, 1H, $J=4.8$), 7.56 (t, 1H, $J=7.6$), 3.03 (t, 2H, $J=6.0$), 2.91 (br.t, 2H), 1.91 (br.m, 4H)
cis-[$PtCl_2$(8-dppn)]	9.86 (d, 1H, $J=5.6$), 8.80 (2d, 1H, $J=4.8$), 8.52 (t, 1H, $J=6.8$), 8.42 (d, 1H, $J=8.8$), 8.15 (t, 1H, $J=6.4$), 8.08 (2d, 1H, $J=7.6$), 7.85 (d, 1H, $J=8.0$), 7.57 (2d, 1H, $J=4.8$), 3.10 (t, 2H, $J=6.8$), 2.94 (t, 2H, $J=6.4$), 2.06 (br.m, 3H), 1.74 (br.m, 3H), 1.52 (br.m, 1H), 1.40 (br.s, 1H)

[a]^{1}H NMR data obtained in DMSO-d_6 at 400 MHz. [b]Ref. [5]. [c]Ref. [9]. d, doublet; 2d, two doublets; dt, doublet of triplet; t, triplet; br., broad; s, singlet; m, multiplet.

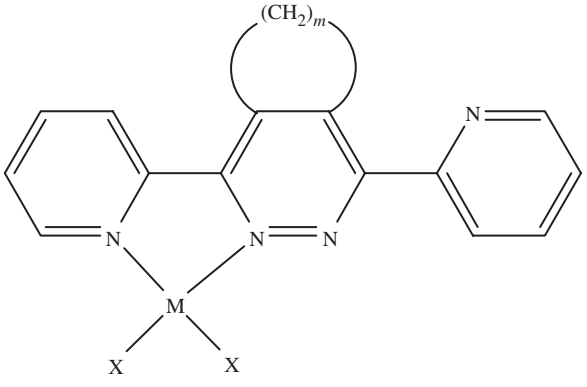


Figure 2. Structure of the complexes: M = Pd; X=Cl; $m=3,4,6,10$; M = Pt; X = Cl ($m=3,4,6$); X = Br ($m=3$).

The highly deshielded protons, $H_{6'}$ and $H_{6'}$, which are directed toward the halide ions show a doublet in the 8.86–9.86 ppm region. These results clearly indicate that the *cis*-dihalo-Pd(II) and Pt(II) with the *n*-dppn ($n=5,6,8$) ligands have the *cis*-configuration [2c, 14a,b, 19] and support the structure shown in figure 2.

Acknowledgments

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