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Ruthenium(0) and Ruthenium(II) Complexes with 1,1,1-Tris(diphenylphosphinomethyl)ethane

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$Ru_3(CO)_{12}$ reacts with the tripodal ligand 1,1,1-tris(diphenylphosphinomethyl)ethane, TDPME, to form the bis-carbonyl complex $Ru(CO)_2$TDPME. Although this new complex does not react with carbon monoxide, it reacts very readily with oxygen to produce the carbonato complex $Ru(CO_3)(CO)$TDPME. Treatment of $Ru(CO)_2$TDPME with chlorine or bromine leads to the initial formation of ionic complexes of the type $[RuX(CO)_2TDPME]X$ which lose carbon monoxide in solution to form neutral Ru(II) complexes: $RuX_2(CO)$TDPME. The complex $RuCl_2(CO)$TDPME apparently may dissociate a phosphine in solution as it reacts with carbon monoxide to form the ionic bis-carbonyl complex isolated previously as an oxidative-addition adduct. The neutral complex reacts reversibly with sulfur dioxide.

In metal carbonyls successive phosphine substitution of CO groups becomes increasingly difficult. For example $Ru(CO)_3(PPh_3)_2$, which has trans phosphine ligands, does not react with excess phosphine although the bis-carbonyl complex $Ru(CO)_2(PPh_3)_3$ has been prepared by an indirect route.[2,3] This effect can be attributed to a stabilization of the remaining M-CO bonds through symbiotic bonding effects and is reflected in the carbonyl stretching frequencies.[4,5] In general, for a given metal and oxidation state, the extent of substitution and the stereochemistry are governed largely by the donor ability and the steric requirements of the bulky phosphines. However, polydentate phosphines may impose substitution patterns and configurations not attainable with monodentate phosphine ligands, and these differences may profoundly influence the chemistry of the metal center. We previously described the effect of the tridentate phosphine **1**, TDPME, with rhodium(I) and iridium(I) complexes.[6] Herein ruthenium(0) and ruthenium(II) complexes derived from **1** are reported.

$$CH_3C(CH_2P(C_6H_5)_2)_3$$

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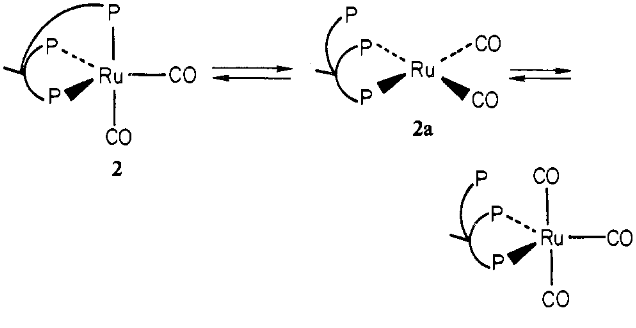
Results and Discussion

When ruthenium carbonyl is treated with the triphosphine **1** and heated in toluene, a new complex is formed. Apparently monomeric with all three phosphines coordinated to ruthenium as in structure **2**, the bright yellow-orange complex exhibits ν_{CO} at 1946 and 1866 cm^{-1} and is extremely sensitive to aerial oxidation, even in the solid state. Because the tripodal phosphine is only capable of occupying three facial sites on a polyhedron, the stereochemistry of **2** must be that with cis carbonyl ligands for which two ν_{CO} would be expected.[6] The low ν_{CO} frequencies are commensurate with carbonyl ligands bound to a ruthenium(0) center which has three "soft" donor ligands.

By contrast, the well-characterized complex $Ru(CO)_3(PPh_3)_2$ having the bulky monodentate phosphines trans exhibits a single ν_{CO} at 1895 cm^{-1} and is relatively air stable in the solid state.[2,7] The tris-substituted carbonyl complex $Ru(CO)_2(PPh_3)_3$ with trans carbonyl ligands and ν_{CO} at 1905 cm^{-1} reacts readily with O_2 in solution to form a stable O_2 adduct.[3] The rapid reaction of $Ru(CO)_2(PPh_3)_3$ with O_2 and other addenda has been interpreted as evidence of phosphine dissociation in solution to form a very reactive four-coordinate species, $Ru(CO)_2(PPh_3)_2$.[3] Similarly, one of the phosphines in **2** may be kinetically labile and the coordinatively unsaturated complex **2a** formed by dissociation of a phosphine from **2** might be expected to react rapidly with oxygen. The unsaturated complex **2a** might also be expected to react with carbon monoxide to form a five-coordinate tris-carbonyl complex in which TDPME functions as a bidentate ligand (Scheme I). The analogous complexes of iron(0), $Fe(CO)_2$TDPME and $Fe(CO)_3$TDPME, have been reported.[8] However, no spectral evidence for $Ru(CO)_3$TDPME was observed when carbon monoxide was bubbled through a toluene solution of **2** or when the preparation of **2** was carried out under 60 psi of carbon monoxide. The failure to observe $Ru(CO)_3$TDPME may be due to a rapid equilibrium which favors $Ru(CO)_2$TDPME, with all three phosphines bound to the metal center, and does not rule out the possibility of a rapid equilibrium between five- and four-coordinate complexes (Scheme I) as was observed earlier in the d^8 rhodium(I) and iridium(I) systems.[6]

Scheme I



Treatment of a solution **2** with oxygen results in the formation of an inert complex characterized as the carbonate **3**. This new complex exhibits a single ν_{CO} at 1998 cm^{-1} and a broad absorption assigned to carbonate at 1600 cm^{-1}. A molecular weight determination confirms that **3** is mono-

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[1] (a) NIH Postdoctoral Fellow, 1969-1971. (b) Visiting Scholar, Stanford University, 1968-1969.

[2] J. P. Collman and W. R. Roper, *J. Amer. Chem. Soc.*, **87**, 4008 (1965).

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[6] W. O. Siegl, S. J. Lapporte, and J. P. Collman, *Inorg. Chem.*, 10, 2158 (1971).

[7] J. Valentine, D. Valentine, Jr., and J. P. Collman, *Inorg. Chem.*, **10**, 219 (1971).

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meric in solution. In a reaction characteristic of metal carbonates containing chelating carbonato ligands,[7,9] complex 3, upon treatment with hydrochloric acid, was converted to the corresponding dichloride 4 (Scheme II). Although $Ru(CO)_3(PPh_3)_2$ reacts slowly with oxygen in solution, possibly *via* a free-radical path, it forms neither a stable carbonate nor an oxygen adduct,[7] whereas $Ru(CO)_2(PPh_3)_3$ reacts rapidly to form a stable O_2 adduct.[3] If $Ru(CO)_2(PPh_3)_3$ and complex 2 both undergo initial dissociation of phosphine and subsequent addition of O_2 to four-coordinate unsaturated complexes, the O_2 adducts thus formed differ in stereochemistry and in the presence of a free phosphine held in close proximity to the metal center of the intermediate derived from 2. By attacking the metal center, the nucleophilic phosphine might assist the rearrangement of an oxygen adduct to the observed carbonato complex 3.[6]

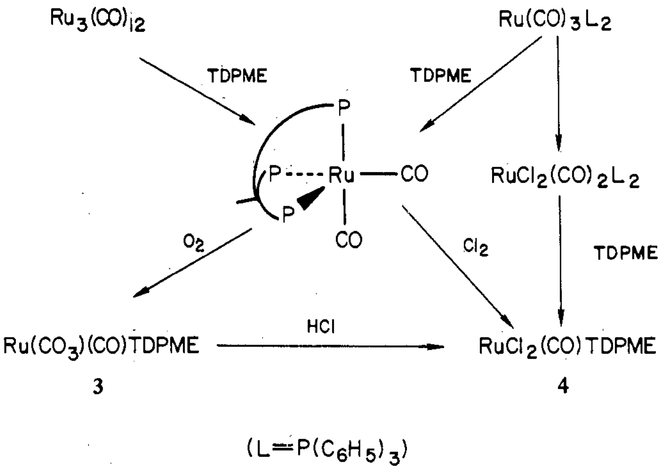
The neutral dichloride complex 4 is monomeric and nonconducting in solution. Because of the TDPME ligand, the stereochemistry of 4 must have CO and the two chlorides cis, giving rise to a single ν_{CO} and two ν_{Ru-Cl} (Table I). Its formulation is further supported by its synthesis *via* two alternate routes (Scheme II). The structure of 4 is noteworthy in that its counterpart with monodentate phosphines, $RuCl_2(CO)L_3$, is known only for highly basic diethylphenylphosphine, in which case the third phosphine could be displaced with carbon monoxide.[10] Where L is triphenylphosphine, $RuCl_2(CO)L_3$ was not isolated; treatment of $RuCl_2(Ph_3P)_3$ with carbon monoxide produced directly the bis-carbonyl complex (eq 1).[11]

$$\mathrm{RuCl_2(PPh_3)_3} \xrightarrow[25^\circ]{\mathrm{CO,\ 1\ atm}} \mathrm{RuCl_2(CO)_2(PPh_3)_2 + PPh_3} \qquad (1)$$

Complex 4 reacted slowly with carbon monoxide at ambient temperature. Bubbling carbon monoxide through a methylene chloride solution of 4 for several minutes produced no change in the infrared spectrum, but stirring the solution for several hours under 40 psi produced a new complex with two carbonyl bands, 2086 and 2048 cm^{-1}. Whereas 4 is a nonelectrolyte, solutions of the bis-carbonyl complex exhibit conductivities in the range observed for 1:1 electrolytes. Thus, the bis-carbonyl complex has been tentatively assigned structure 6 (Scheme III).

It appears likely that one of the phosphine groups of 4 is also labile and in the presence of carbon monoxide the vacant coordination site is filled by a molecule of carbon monoxide to form initially $RuCl_2(CO)_2TDPME$, 5 (Scheme III). Such a process is reasonable; the iron(II) analog of 5 is known[12] and $RuCl_2(CO)(P(C_2H_5)_2Ph)_3$ and $RuCl_2(CO)(PPh_3)_3$ (the hypothetical intermediate in reaction 1) both undergo rapid replacement of a phosphine by carbon monoxide.[10,11] Although with $RuCl_2(CO)_2(Ph_3P)$ in the presence of excess triphenylphosphine, the displacement of chloride by the free phosphine of complex 5, to form the observed product 6, may be favored because it is intramolecular. Similar intramolecular displacements of chloride by an uncoordinated phosphine of TDPME were observed earlier with rhodium(I) and iridium(I) complexes.[6] Complex 6 was isolated but rapidly lost carbon monoxide at room temperature and was characterized as the hexafluorophosphate salt.

Scheme II



Scheme III

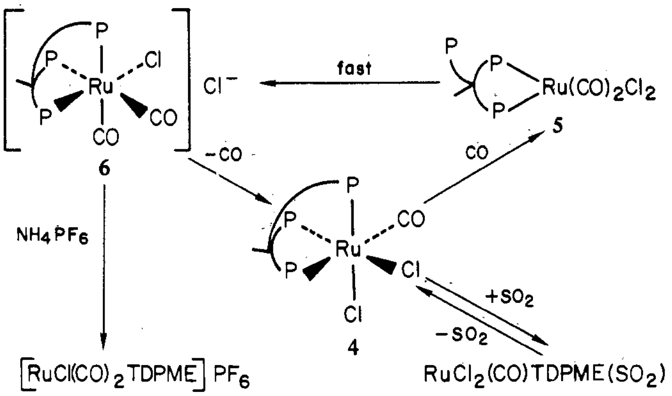


Table I. Infrared Data (cm^{-1})[a]

Compd	Freq: Carbonyl str	Freq: Other
$Ru(CO)_2(TDPME)$[b]	1946 vs, 1866 s	
$Ru(CO_3)(CO)(TDPME)$	1998 s	ν_{CO_3} 1600 br, w
$RuCl_2(CO)(TDPME)$	2010 s	ν_{Ru-Cl} 289 w, 265 w
$[RuCl(CO)_2(TDPME)]^+Cl^-$	2078 vs, 2038 s	
$[RuCl(CO)_2(TDPME)]^+PF_6^-$	2076 vs, 2040 s	
$[RuBr(CO)_2(TDPME)]^+Br^-$	2076 s, 2036 s	
$RuBr_2(CO)(TDPME)$	2008 s	

[a] For spectra run in KBr unless noted otherwise. [b] THF solution.

When a solution of 4 was saturated with sulfur dioxide, the ν_{CO} in 4 diminished and a new band appeared at higher frequency (2078 cm^{-1}); purging the solution with nitrogen resulted in the reappearance of complex 4. The sulfur dioxide adduct was not sufficiently stable to allow its isolation. The large shift in ν_{CO} observed upon addition of sulfur dioxide to 4 is in accord with the shift expected if SO_2 is complexed with the metal center rather than to a halide.[13] It is likely that SO_2 adduct formation involves prior dissociation of a phosphine and complexation of SO_2 with a coordinatively unsaturated ruthenium(II) complex since reaction of SO_2 with saturated d^6 complexes is rare.

The oxidative addition of halogens to the d^8 Ru(0) complex 2 occurred stepwise with initial formation of ionic halide salts (eq 2). Addition of excess chlorine to a toluene solution 2 at 25° resulted in immediate precipitation of a yellow powder which partially decomposed on warming.

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(9) K. R. Laing and W. R. Roper, *Chem. Commun.*, 1568 (1968).

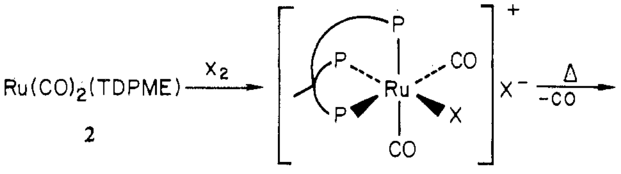
(10) J. Chatt, B. L. Shaw, and A. E. Field, *J. Chem. Soc.*, 3466 (1964).

(11) T. A. Stephenson and G. Wilkinson, *J. Inorg. Nucl. Chem.*, **28**, 945 (1966).

(12) H. Behrens, E. Linder, and H. D. Feilner, *Z. Anorg. Allg. Chem.*, **385**, 325 (1971).

(13) M. R. Snow, J. McDonald, F. Basolo, and J. A. Ibers, *J. Amer. Chem. Soc.*, **94**, 2526 (1972).

The yellow powder exhibited two carbonyl stretching bands in the infrared spectrum (Table I) and was spectroscopically identical with the cationic bis-carbonyl complex **6** described above. The cationic complex was stable when isolated as its hexafluorophosphate salt. The chloride salt, when heated in tetrahydrofuran solution was converted to the previously isolated neutral dichloride complex **4** with concomitant loss of carbon monoxide (eq 2). Addition of bromine to complex **2** resulted in the immediate precipitation of a yellow powder which also exhibited two strong carbonyl bands in the infrared spectrum (Table I), suggesting formation of the ionic bromide salt **7**. Analysis of **7** indicated that it remained impure despite several washings; attempts to purify the ionic bromide by recrystallization resulted in its conversion to the neutral dibromide **8**.



6, X = Cl
7, X = Br

$RuX_2(CO)(TDPME)$ (2)

4, X = Cl
8, X = Br

Complex **2** underwent oxidative addition reactions with other addenda (*e.g.*, CH_3SO_2Cl) in a similar stepwise manner. Adducts obtained were characterized spectroscopically but analytical samples were difficult to obtain. A reasonable supposition would be that in certain cases the phosphine which can dissociate functions as a second nucleophilic site in the complex, allowing the formation of hard to separate bis adducts.

Conclusion

Ruthenium(0) and ruthenium(II) carbonyl complexes with the tripodal ligand TDPME exhibit degrees of phosphine substitution and stereochemistries not generally obtained with monodentate phosphines of similar basicity. The unique chemical reactivity of these TDPME complexes might be explained by the presence of a labile phosphine which allows formation of highly reactive coordinatively unsaturated complexes. The chemistry of adducts formed *via* such unsaturated intermediates may be influenced by the uncoordinated phosphine which is held near the metal center and may provide nucleophilic assistance to intramolecular rearrangement. Thus reactions are observed which are not common to analogous complexes containing monodentate phosphines.

Experimental Section

General Information. Microanalyses and molecular weight determinations (osmometric) were performed by Messrs. E. Meier and J. Consul, Stanford Microanalysis Laboratory. Infrared spectra were recorded on a Perkin-Elmer Model 457 spectrometer; only selected ir bands have been reported. Conductivity measurements were made with a Yellow Springs Instrument Model 31 conductivity bridge in 10^{-3} M methylene chloride solution.

Preparative reactions were carried out under argon using Schlenk-tube techniques. All solvents were degassed prior to their use. The tridentate ligand TDPME was prepared by the published procedure,[14] as were the complexes $Ru(CO)_3(P(C_6H_5)_3)_2$ and $RuCl_2(CO)_2(P(C_6H_5)_3)_2$.[2]

Preparation of $Ru(CO)_2$(TDPME). (A) A solution of 0.24 mmol of TDPME and 0.08 mmol of $Ru_3(CO)_{12}$ in 10 ml of toluene was heated with the oil bath temperature at 130° for 3-4 hr. The initial orange color of the solution went to dark red to orange-red. Hexane (*ca.* 15 ml) was added to precipitate the product. The yellow-orange product was collected by filtration, washed with *n*-hexane, and dried. It was impossible to handle the solid in the atmosphere without decomposition.

(B) A solution of $Ru(CO)_2$(TDPME), spectroscopically identical with the toluene solution above, was obtained when 0.1 mmol of $Ru(CO)_3(P(C_6H_5)_3)_2$ and 0.1 mmol of TDPME were heated in 15 ml of toluene for 5 hr with an oil bath at 120°. The product was treated with oxygen and isolated as the carbonate (see below).

Anal. Calcd for $C_{43}H_{39}O_2P_3Ru$: C, 66.06; H, 5.03. Found: C, 65.93; H, 5.19.

Preparation of $Ru(CO_3)(CO)$(TDPME). (A) A solution of 0.1 mmol of $Ru(CO)_3(P(C_6H_5)_3)_2$ and 0.1 mmol of TDPME in 15 ml of toluene was heated with the oil bath at 120° for 5 hr. After cooling, oxygen was bubbled through the solution for 5 min and the solvent was evaporated *in vacuo*. The residue was recrystallized from CH_2Cl_2-*n*-hexane; 33% of the white crystalline carbonate was obtained.

(B) Air was bubbled through a solution of $Ru(CO)_2$(TDPME), prepared *in situ* from TDPME and $Ru_3(CO)_{12}$ as described above. The solvent was evaporated *in vacuo* and the residue crystallized from CH_2Cl_2-*n*-hexane to afford 22% of the white crystalline carbonate.

Anal. Calcd for $C_{43}H_{39}P_3O_4Ru$: C, 63.46; H, 4.83; mol wt 814. Found: C, 61.06; H, 4.78; mol wt 905 (CH_2Cl_2).

Preparation of $RuCl_2(CO)$(TDPME). (A) A solution of 0.1 mmol of TDPME and 0.1 mmol of $RuCl_2(CO)_2(P(C_6H_5)_3)_2$ in 10 ml of toluene was heated with the oil bath at 120° for 6 hr. A yellow powder deposited and was collected by filtration, washed with *n*-hexane, and dried. A 78% yield of the desired dichloride was obtained.

(B) Dry HCl gas was bubbled in a solution of 0.05 mmol of $Ru(CO)_3(CO)$(TDPME) in 5 ml of CH_2Cl_2. The yellow solution darkened initially and then became pale yellow. After stirring for 10 min at room temperature, *n*-hexane was added to the cloud point; cooling at 0° gave pale yellow crystals which were washed with *n*-hexane and dried. The yield was 68%.

(C) Chlorine gas bubbled into a toluene solution of 0.24 mmol of $Ru(CO)_2$(TDPME) which had been prepared *in situ* and then cooled to −25°. A yellow solid deposited immediately. The mixture was allowed to warm to room temperature and the solvent was removed *in vacuo*. THF (15 ml) was added to the residue and the solution was refluxed for 30 min. After the mixture had cooled, it was filtered. The yellow powder was washed with ether and with hexane and then dried to yield 46% of the neutral dichloride. A second crop (32%) of less pure material was obtained from the filtrate.

Anal. Calcd for $C_{42}H_{39}Cl_2O_3P_3Ru$: C, 61.17; H, 4.77; Cl, 8.60; mol wt 824. Found: C, 60.72; H, 5.06; Cl, 8.92; mol wt 793 (CH_2Cl_2).

Preparation of $[RuBr(CO)_2TDPME]^+Br^-$. A solution of 0.24 mmol of $Ru(CO)_2$(TDPME) prepared *in situ* in 10 ml of toluene was cooled to −25°. Bromine (0.24 mmol) was added dropwise and a yellow precipitate deposited immediately. After being stirred 15 min, the mixture was filtered and the product was washed with ether and dried. The pale yellow powder represented an 87% yield. Attempts to recrystallize the product resulted in formation of the neutral dibromide $RuBr_2(CO)$(TDPME) (see below).

Anal. Calcd for $C_{43}H_{39}O_2Br_2P_3Ru$: C, 54.85; H, 4.17; Br, 17.0. Found: C, 52.83; H, 4.27; Br, 18.16.

Preparation of $RuBr_2(CO)$(TDPME). A solution of 0.24 mmol of $Ru(CO)_2$(TDPME), prepared *in situ* in 10 ml of toluene, was cooled to −25°. Bromine (0.24 mmol) was added dropwise and the mixture was allowed to warm to room temperature. The solvent was removed *in vacuo*. THF (12 ml) was added to the residue and this mixture was stirred at 55° for 30 min. After cooling, the yellow-green product was collected by filtration, washed with ether, and dried; a yield of 51% was obtained.

Anal. Calcd for $C_{42}H_{39}OBr_2P_3Ru$: C, 55.25; H, 4.03; Br, 17.49. Found: C, 54.00; H, 4.64; Br, 16.92.

Reaction of $RuCl_2(CO)$TDPME with CO. When a methylene chloride solution of $RuCl_2(CO)$TDPME was stirred under 10 psi of CO and sampled after *ca.* 1 hr, the infrared spectrum contained two new ν_{CO} at 2086 and 2046 cm^{-1}. When stirred under 40 psi of CO for several hours, the solution no longer exhibited ν_{CO} for starting material; only the two new bands were observed. If nitrogen was then bubbled through the solution or the solution was heated to reflux in the absence of CO, starting material was recovered. Conductance measurements recorded on the solution containing the bis-

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[14] W. Hewertson and H. R. Watson, *J. Chem. Soc.*, 1490 (1962).

carbonyl were in the range recorded for model 1:1 electrolytes,[15] whereas measurements on the initial solution indicated the starting material was a nonelectrolyte.

Isolation of [RuCl(CO)$_2$TDPME]PF$_6$. A suspension of 111 mg of RuCl$_2$(CO)TDPME and 40 mg (an excess) of NH$_4$PF$_6$ in 8 ml of acetone was stirred under 40 psi of CO for 4 hr. The suspension color changed from yellow to white. Water was added dropwise, yielding initially a colorless solution and then a white precipitate. The solid was collected, washed with water, and dried to yield 105 mg of white powder. The infrared spectrum contained a strong band at 835 cm^{-1} characteristic of PF$_6^-$ as well as the two ν_{CO}.

Anal. Calcd for $C_{43}H_{39}ClO_2P_4F_6Ru$: C, 53.68; H, 4.09; Cl, 3.68. Found: C, 54.71; H, 5.12; Cl, 3.23.

Registry No. Ru(CO)$_2$(TDPME), 37843-33-7; Ru(CO$_3$)(CO)(TDPME), 37843-34-8; RuCl$_2$(CO)(TDPME), 37843-35-9; [RuCl(CO)$_2$(TDPME)]Cl, 37843-36-0; [RuCl(CO)$_2$(TDPME)]PF$_6$, 37843-37-1; [RuBr(CO)$_2$(TDPME)]Br, 37897-34-0; RuBr$_2$(CO)(TDPME), 37843-38-2; TDPME, 22031-12-5; Ru, 7440-18-8; SO$_2$, 7446-09-5.

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Crystal and Molecular Structure of Bis(2,4-pentanedionato)dimethyltin(IV)[1]

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The crystal structure of bis(2,4-pentanedionato)dimethyltin(IV), $(CH_3)_2Sn(C_5H_7O_2)_2$, has been determined from three-dimensional X-ray diffraction data. Unit cell dimensions are $a = 7.12 \pm 0.01$ Å, $b = 13.87 \pm 0.02$ Å, $c = 7.69 \pm 0.01$ Å, and $\beta = 104.7 \pm 0.2°$. The monoclinic space group is $P2_1/n$. The density by flotation is 1.59 g/cm^3. Two molecules in the unit cell give a calculated density of 1.57 g/cm^3. By visual estimation of film data, the intensities of 1195 independent reflections were obtained. The structure was solved by three-dimensional Patterson and Fourier methods and refined by full-matrix least-squares to a final conventional R factor of 7.9%. The arrangement of the atoms about the tin forms a nearly perfect octahedron. The 2,4-pentanedionate acts as a bidentate ligand bonding to the tin atom through the oxygen atoms. The bond distances in the octahedron are as follows: Sn–C(3), 2.14 (2) Å; Sn–O(1), 2.20 (1) Å; Sn–O(2), 2.18 (1) Å. The space group and the presence of only two molecules in the unit cell require the tin atom to be on a center of symmetry, and therefore the two methyl groups bonded to the tin must be in trans positions.

Introduction

The crystal structure of the bis(2,4-pentanedionato)dimethyltin(IV) molecule has been determined to resolve some of the controversy that has occurred over the structure of this compound in the literature. A literature survey shows that until 1969, most authors felt that the methyl groups should be in the trans positions. The techniques used in those studies were Raman, infrared, nmr, and Mossbauer.[4-7] In 1969, Moore and Nelson,[8] on the basis of a dipole moment study, assigned the compound the cis structure.

In the work of McGrady and Tobias[6] the infrared and Raman spectra were used to compare the structure of the compound in solution to that in the solid. If the methyl groups were in the trans position, then the skeletal symmetrical stretching vibrations should be only Raman active with the antisymmetrical skeletal vibrations being only infrared active. If the methyl groups were in the cis positions, then all skeletal vibrations would be Raman and infrared active. The observed bands were in keeping with a trans structure in both the solid and solution. However, it was noted that the symmetrical C-O vibration which should have been Raman active was absent.

From the dipole moment values obtained in benzene (2.95 D) and cyclohexane (3.19 D) Moore and Nelson[8] assigned the cis structure to the compound since the trans structure should have a dipole moment of zero. They felt that a cis type structure with some distortion from a true octahedron would equally well fit the accumulated data.

Experimental Section

Crystal Preparation. The crystals were prepared by the use of a slight modification of methods previously published.[6,8] Dimethyltin oxide (2.75 g, 0.02 mol) was added to 10 ml of 2,4-pentanedione and refluxed for 5.5 hr. The clear solution was decanted and kept at 0° overnight. The colorless crystals were then dissolved in hot benzene containing a small amount of 2,4-pentanedione and recrystallized; mp 177–179°. *Anal.* Calcd for $C_{12}H_{20}O_4Sn$: C, 41.54; H, 5.81. Found: C, 41.63; H, 5.82.

X-Ray Data. Bis(2,4-pentanedionato)dimethyltin(IV) crystallizes in the monoclinic system with $a = 7.12 \pm 0.01$ Å, $b = 13.87 \pm 0.02$ Å, $c = 7.69 \pm 0.01$ Å, and $\beta = 104.7 \pm 0.2°$. A flotation measurement in aqueous KI solution gave a density of 1.59 ± 0.01 g/cm^3. A calculated density of 1.569 ± 0.005 g/cm^3 is obtained assuming two molecules per unit cell. The systematic extinctions require that on

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[1] An initial report of this work was made at the 5th Midwest Regional Meeting of the American Chemical Society, Kansas City, Mo., Oct 29-31, 1969; see Abstracts, No. 319.

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