

Table I. T_1 Values of Hydrido Ligands in the Compounds Studied

compd	no.	T_1 , s	temp, °C
$\text{H}_2\text{Os}_3(\text{CO})_{10}$	1	6.5	+20
		1.0	-60
$\text{HDO}_3(\text{CO})_{10}$	2	30.2	+20
$\text{H}_2\text{Os}_3(\text{CO})_{11}$	3	0.9 (terminal)	-60
		0.9 (bridging)	-60
$\text{H}_4\text{Ru}_4(\text{CO})_{12}$	4	4.8	+20
		1.4	-30
$\text{H}_4\text{Ru}_4(\text{CO})_{11}\text{PPh}_3$	5	1.6	+20
		0.8	-30
$\text{H}_4\text{Ru}_4(\text{CO})_9[\text{P}(\text{OEt})_3]_3$	6	1.2	+20
$\text{HRu}_3(\text{CO})_9(\text{MeCCHCMe})$	7	24.1	+20
$\text{H}_2\text{Ru}_3(\text{CO})_9(\text{MeC}=\text{CEt})$	8	1.0	-60
$\text{H}_2\text{Os}_3(\text{CO})_9(\text{C}=\text{CH}_2)$	9	4.3	+20

metal-metal site below the Ru_3 triangle opposite to the σ/π -bonded allylic chain.

In principle, if T_1^{DD} between two ^1H nuclei can be accurately determined (through measurement of the nuclear overhauser enhancement when the two ^1H nuclei resonate at different frequencies) and the molecular correlation time τ_c is known by an independent experiment, $^1\text{H}-T_1$ can provide a method to evaluate the internuclear distance between the two interacting nuclei. However when the dipolar interaction involves two magnetically equivalent nuclei as in **1**, T_1^{DD} can be obtained by a different approach. This consists in measuring the $^1\text{H}-T_1$ of the monodeuteriated species $\text{HDO}_3(\text{CO})_{10}$ (**2**); its T_1 is definitely long than that found for $\text{H}_2\text{Os}_3(\text{CO})_{10}$ (**1**), and their difference allows an accurate evaluation of the dipole-dipole relaxation rate between the two equivalent hydrides⁴

$$R^{\text{DD}} = \frac{1}{T_1^{\text{DD}}} = \left\{ \frac{1}{T_1(1)} - \frac{1}{T_1(2)} \right\} \cdot \frac{1}{0.96} \quad [\text{s}^{-1}]$$

Now, applying the usual equation⁵ for $1/T_1^{\text{DD}}$ for the intramolecular $^1\text{H}-^1\text{H}$ interaction in the fast correlation time limit, we get

$$r_{\text{H-H}} = \sqrt[6]{\frac{3}{2} \gamma_{\text{H}}^4 \hbar^2 \tau_c T_1^{\text{DD}}}$$

and using a τ_c value of 25 ps (which proved to be satisfactory in the study of ^{13}C and ^{17}O relaxation behavior in the same molecule⁶), we have calculated a $r_{\text{H-H}}$ distance of $2.35 \pm 0.03 \text{ \AA}$. This distance is very similar to that found in the solid state from a neutron diffraction study (2.37 \AA).⁷

An analogous approach could be used to calculate the M-H distance if different metal isotopes are available. This is the case of the $[\text{Pt}(\text{PET}_3)_3\text{H}]\text{BF}_4$ complex reported in ref 2, for which the hydride- T_1 measurement for the ^{195}Pt isotopomer ($I = 1/2$) is slightly shorter (2.03 s) than the corresponding T_1 in the isotopomer having $I = 0$ platinum nuclei (2.38 s). The dipolar contribution to the hydride relaxation rate due to the magnetically active ^{195}Pt is then equal to 0.0724 s^{-1} ; assuming $\tau_c = 60 \text{ ps}$ (reasonable value for ionic mononuclear complex), a Pt-H distance of 1.67 \AA would be obtained.

Experimental Section

Compounds **1**,⁸ **3**,⁸ **4**,⁹ **5**,⁹ **6**,⁹ **7**,³ **8**,¹⁰ and **9**¹¹ were obtained by litera-

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ture methods; their purity was checked by the usual spectroscopic techniques, i.e. MS, IR, and ^1H NMR.

Compound **2** was obtained by reacting $\text{Os}_3(\text{CO})_{12}$ (150 mg, 0.165 mmol) (**1**) with D_2 (4.43 mmol) in a sealed vial at $+110 \text{ }^\circ\text{C}$ for 12h; after TLC workup, a mixture of the fully deuterated species $\text{D}_2\text{Os}_3(\text{CO})_{10}$ ^{12,13} and the monodeuteriated derivatives $\text{HDO}_3(\text{CO})_{10}$ (**2**) was achieved in a 9:1 molecular ratio. Since ^1H NMR cannot discriminate between **1** and **2**, the identity of **2** was verified from the observation of the isotopic shift of the two equivalent CO groups trans to the metal-metal bonds on the osmium atoms bridged by the hydrido ligands. These resonances fall at 176.085, 175.925, and 175.765 ppm in **1**, **2**, and $\text{D}_2\text{Os}_3(\text{CO})_{10}$, respectively.

NMR measurements were carried out on a JEOL GX-270/89 spectrometer. The samples were prepared by using Schlenk tube techniques and oxygen-free CDCl_3 . The nonselective inversion-recovery pulse sequence was used to obtain T_1 values. Errors are estimated to be $\pm 5\%$.

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Registry No. $\text{H}_2\text{Os}_3(\text{CO})_{10}$, 41766-80-7; $\text{HDO}_3(\text{CO})_{10}$, 81293-87-0; $\text{H}_2\text{Os}_3(\text{CO})_{11}$, 56398-24-4; $\text{H}_4\text{Ru}_4(\text{CO})_{12}$, $\text{H}_4\text{Ru}_4(\text{CO})_{12}$; $\text{H}_4\text{Ru}_4(\text{CO})_{11}\text{PPh}_3$, 34742-79-5; $\text{H}_4\text{Ru}_4(\text{CO})_9[\text{P}(\text{OEt})_3]_3$, 72950-52-8; $\text{HRu}_3(\text{CO})_9(\text{MeCCHCMe})$, 56943-13-6; $\text{H}_2\text{Ru}_3(\text{CO})_9(\text{MeC}=\text{CEt})$, 108189-63-5; $\text{H}_2\text{Os}_3(\text{CO})_9(\text{C}=\text{CH}_2)$, 42765-74-2.

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Contribution from the Department of Chemistry,
University of British Columbia, Vancouver,
British Columbia, Canada V6T 1Y6

High-Resolution Solid-State ^{13}C NMR Study of Bis(2,4-pentanedionato)zinc Complexes

K. Takegoshi, K. J. Schenk, and C. A. McDowell*

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High-resolution solid-state ^{13}C NMR spectroscopy,¹ involving the combined techniques² of cross-polarization (CP)³ and magic-angle spinning (MAS),⁴ and X-ray powder diffraction were used to study anhydrous (monomeric) (**1**), hydrated (**2**), and trimeric (**3**) bis(2,4-pentanedionato)zinc. The results were interpreted, in combination with published structural information, to derive a correlation between the geometry of the carbonyl groups and the delocalization of the π electrons in the acetylacetonate ligands. A novel metastable phase, and its phase behavior, was also investigated.

A early crystal structure of **2** is available.⁵ The zinc atom is coordinated by a distorted tetragonal ZnO_5 pyramid, and the intramolecular local configurations around the four carbonyl groups in **2** are identical within $\pm 2^\circ$ in angle and $\pm 0.04 \text{ \AA}$ in bond length. We therefore expect the π electrons to be localized. Only

* To whom correspondence should be addressed.

the molecular structure of **1** in the gas phase is known.⁶ The observed D_{2d} symmetry of the ZnO_4 skeleton was well reproduced by recent ab initio calculations.⁷ These also indicate the complete delocalization of the π electrons in **1** in the gas phase. The crystal structure of **3** has been determined accurately;⁸ but since some of the carbonyl groups are bridging, their chemical shifts cannot straightforwardly be related to the degree of delocalization of the π electrons.

We show in this paper that the isotropic chemical shift values of the carbonyl ^{13}C nuclei in metal-acetylacetonate chelate (M-acac) complexes can complement structural information in assessing the degree of delocalization of the π electrons in M-acac complexes.

Experimental Section

The monohydrate, **2**, was prepared by following a well-proven synthetic route.⁹ It should be noted that the prismatic, transparent needles of **2** are not very stable even at room temperature, as evidenced by the crystal faces becoming opaque after several days. Compound **2** can be converted to **1** by drying the former in vacuo over P_2O_5 for about 1 day. Water removal by heating seems to result in the same phase.¹⁰ The monomer, **1**, can be sublimated in vacuo (0.5 Torr) at about 60 °C to yield **3**.⁸ Compounds **1** and **3** represent different modifications of anhydrous Zn-acac, the former being metastable and transforming into the latter in vacuo. The existence of these two phases has been demonstrated by means of ^{13}C CP-MAS NMR spectra and X-ray diffraction. A Debye-Scherrer photograph ($Cu K\alpha$) taken immediately after drying **2** (powder inside a capillary tube) contains some prominent lines ($d = 7.1 \text{ \AA}$ and $d = 6.5 \text{ \AA}$), which cannot be accounted for by **2** and/or **3**; however, several weak lines of **2**⁵ and **3**⁹ are observed.

White $Cd(acac)_2$ (**5**) was prepared by following the synthetic route which worked well for **2**; another specimen of that compound was also purchased from K&K laboratories, Plainview, NY. Both samples did not yield the expected values in a microanalysis (Anal. Calcd: C, 38.7; H, 4.5. Found: C, 36.1–37.7; H, 4.2–4.4). The red $Cd(acac)_2$ could not be prepared by following the instructions given in ref 19. A Debye-Scherrer X-ray photograph of the white compound was taken by using $Cu K\alpha$ radiation. The well-resolved lines up to $\theta = 13^\circ$ can all be indexed by using the unit cell given for the reddish compound.²⁰

Measurements of ^{13}C and ^{113}Cd CP-MAS NMR spectra were performed on a Bruker CXP200 FT NMR spectrometer operating at 200.05 MHz for 1H , 50.3 MHz for ^{13}C , and 44.36 MHz for ^{113}Cd . The rf field strength for 1H , ^{13}C , and ^{113}Cd was about 42 kHz. An Andrew-Beams type deuterated Plexiglas rotor was used for the MAS at a spinning frequency of approximately 3.0 kHz. The setting of the magic angle was monitored by the ^{79}Br NMR spectrum of KBr incorporated in the rotor.¹¹ FID signals were accumulated 200–500 times with a contact time of 2 ms and a repetition time of 4 s. To increase the efficiency of the experiments, the 1H magnetization remaining after a decoupling period was flipped back to an axis parallel to the static magnetic field applying a 90° pulse.¹² The ^{13}C chemical shifts were calibrated in ppm relative to Me_4Si as previously described.¹³ The ^{113}Cd chemical shifts were calibrated in ppm relative to a 0.1 M aqueous $Cd(ClO_4)_2$ solution of ionic strength $I = 4.5$ by taking the chemical shifts of solid $3CdSO_4 \cdot 8H_2O$ as an external reference.²¹

Results and Discussion

Wilkie and Haworth¹⁴ observed a singlet ^{13}C NMR signal for the carbonyl carbon nuclei of many M-acac complexes in solution

Table I. Isotropic ^{13}C Chemical Shifts for Carbon Nuclei in **1**, **2**, **3**, and **5**^a

carbon nucleus	Zn-acac			Cd-acac	
	in solids		in $CDCl_3$	in solids	in $CDCl_3$
	1	2	3	5	ref 14
C=O			199.2		
		200.6	197.6	199.7	
	195.7	194.5	194.0	194.8	194.3
		192.0	191.3		190.3
		189.0	190.3		187.6
CH		101.7	103.2		
	102.5	100.6	101.9	100.8	101.0
			99.4		97.7
CH ₃			30.4		
	30.2	29.2	29.3		30.8
	28.6	28.0	27.9	28.1	30.4
		27.4	27.4		29.9
			27.0		

^aChemical shifts are in ppm from Me_4Si as a standard.

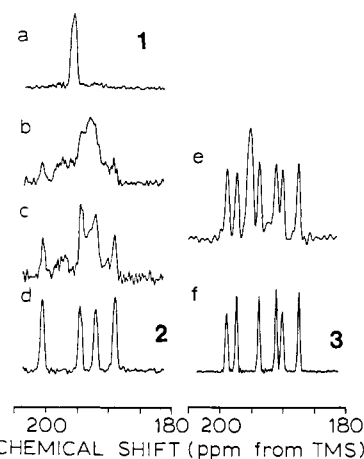


Figure 1. ^{13}C CP-MAS spectra for **1** (a), **2** (d), and **3** (f). The remaining spectra show the changes resulting from storing **1** in moist air [(b) 4 h; (c) 12 h] and in vacuo [(e) for 2d].

with $\delta = 186$ – 195 , depending on the solvents and the metal nucleus. At first sight, the observation of a singlet for the two carbonyl carbon nuclei of the compound in solution seems to be good evidence that the two carbonyl carbon nuclei in M-acac complexes are equivalent and that the π electrons are completely delocalized. However, a molecule in solution is free from crystal packing forces, and a fast alternation of the metal-oxygen coordination may occur to average out the chemical shift differences between the carbonyl carbon nuclei.

The direct correspondence between the chemical shifts in solid **1** and those in $CDCl_3$ (Table I) is noteworthy; the singlet ^{13}C NMR signal at $\delta = 195.7$ in **1** clearly demonstrates that the four carbonyl carbon nuclei are *equivalent*, indicating almost complete delocalization of the π electrons within the two acac residues of **1** in the solid state. This result suggests a highly symmetric coordination of the zinc atom. If one takes into account the molecular structure of **1** in the gas phase,⁷ it appears, therefore, reasonable to postulate that **1**, obtained as described above, should be monomeric Zn-acac adopting approximately a D_{2d} conformation. The slight asymmetry of the line (Figure 1a) could be interpreted as indicating a distortion from the regular tetrahedral geometry either due to crystal packing forces, i.e. the crystalline field, or by an incomplete removal of water in forming the anhydrous compound. This hypothesis is further sustained by the strict $\bar{4}2m$ symmetry of the zinc coordination in bis(2,2,6,6-tetramethyl-3,5-heptanedionato)zinc.²²

In **2**, the well-resolved four singlet ^{13}C NMR signals appearing at $\delta = 188$ – 200 indicate that the four carbonyl carbon nuclei are *inequivalent*. A series (Figure 1a–d) of spectra show the changes

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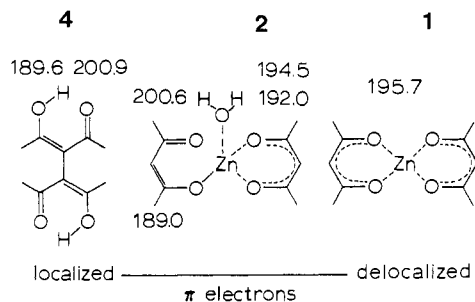


Figure 2. Schematic representations of the relations between the ^{13}C chemical shifts for the carbonyl carbon nuclei and the delocalization of the π electrons in compounds **1**, **2**, and **4**.

in the spectrum of **1** upon absorbing moisture from the air. Clearly, an additional coordination of a water molecule into the ZnO_4 structure in **1** perturbs the electronic energy levels of the oxygen atoms in the acac residues, leading to inequivalent ^{13}C chemical shifts in **2**. Thus, this inequivalence of the ^{13}C chemical shifts can be attributed to incomplete delocalization of the π electrons in the acac residues. The chemical shifts of the carbonyl carbon nuclei are, therefore, a more sensitive probe for studying delocalization of the π electrons in acac residues than the atomic coordinates from signal-crystal x-ray diffraction determinations; for in **2** the latter method predicts almost equivalent local structures for the four carbonyl groups.⁵ Some lines with small intensities in the NMR spectra shown in Figure 1b,c, which cannot be assigned to either **1** or **2**, may be attributed to other modifications of the Zn-acac complex, such as **3**.

The chemical shifts of the four different carbonyl carbon nuclei in **2** can be assigned as follows. The close resemblance of the chemical shift values of 200.6 and 189.0 ppm in **2** to those of 200.9 and 189.6 ppm in 3,4-diacetyl-2,5-hexanedione (**4**)¹⁵ lead us to conclude that those two carbon nuclei in **2** are in one acac residue in which the degree of delocalization of the π electrons is similar to that in the acac residue in **4**. This does, however, not mean that the π -electrons are completely localized in one of the two acac residues in **2**. On the contrary, in the interpretation of both the diffraction¹⁶ and solid-state NMR¹⁵ studies, it was postulated that delocalization of the π electrons occurs in **4**. The other two chemical shifts of similar value, namely 192.0 and 194.5 ppm, in **2** can be assigned to the other acac residue, the π electrons of which are *more* completely delocalized. Figure 2 summarizes these conclusions.

Another series of spectra (Figure 1a,e,f) show that **1** gradually transforms into **3** in vacuo; i.e., the most stable form of anhydrous Zn-acac is the trimer. There are six independent carbonyl carbon nuclei in a trimer (Figure 3), four of which are "chelating only" and two of which are "chelating and bridging". Thus, the six ^{13}C CP-MAS signals in Figure 1f can be assigned to the six *chemically inequivalent* carbonyl carbon nuclei in the molecule. Further, it is interesting to note that the CP-MAS spectrum of **3**, obtained by sublimation of **1** (Figure 1f), shows narrower lines than that of **3**, obtained by storing **1** in vacuo for 2 days (Figure 1e). The broader line width found in the latter trimeric complex may be due to crystal imperfections.

To investigate the possibility of an exchange of the four inequivalent carbonyl groups in **2** by alternation of the π bondings, two-dimensional exchange NMR experiments¹⁷ were performed for the four ^{13}C resonances of the carbonyl carbon nuclei in **2**. However, no "cross peaks" could be observed in our experiments with mixing times up to 3 s at room temperature. This implies that the π electrons are *localized* in **2**.

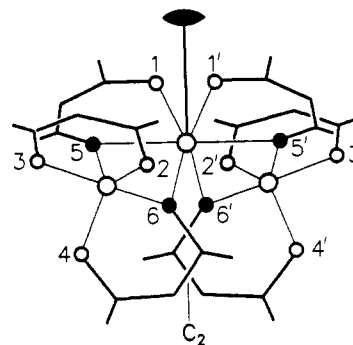


Figure 3. Schematic structure of **3**. The chelating-only oxygen atoms are denoted by the symbol \circ , the chelating-bridging oxygen atoms by \bullet , and the zinc atoms by \bigcirc . The thick lines represent the carbon skeleton, and the thin lines the oxygen-zinc coordination. C_2 and the solid ellipsoid indicate the twofold rotation axis within a molecule.

$\text{Ce}(\text{acac})_2$ has been reported to exist in a white¹⁸ and a reddish¹⁹ modification, both of which do not contain any water molecules. A tetrahedral coordination of the carbonyl groups is therefore to be expected. CP-MAS ^{13}C NMR spectra should then help us further to substantiate or to reject our hypothesis concerning the delocalization of the π electrons in **1**. Both samples of white $\text{Cd}(\text{acac})_2$ studied gave rise to the same spectrum; unfortunately, it was different from the one of monomeric $\text{Zn}(\text{acac})_2$. The chemical shift values are summarized in Table I. This surprising result can be understood by recalling that the structure of the white $\text{Cd}(\text{acac})_2$ seems to be identical with that of reddish $\text{Cd}(\text{acac})_2$ determined by X-rays²⁰ and that the slight color of the reddish compound may be due to impurities, such as CdO . This structure consists of edge-sharing Cd, O -octahedra that form linear polymers. These chains contain four independent carbonyl carbon nuclei and one independent Cd nucleus. All the structural features can be explained convincingly by our observation of the four carbonyl lines in the ^{13}C spectrum, the one line in the ^{113}Cd spectrum, and its chemical shift value (~ 18 ppm), which is characteristic of an octahedral coordination around the Cd atom.²¹

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Contribution from Rocketdyne, A Division of Rockwell International, Canoga Park, California 91303

Solid-Propellant-Based Pure Fluorine Gas Generators

Karl O. Christe* and Richard D. Wilson

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The storage and handling of either cryogenic liquid fluorine or high-pressure gaseous fluorine frequently presents safety and logistics problems. These problems can be overcome by the use of solid-propellant fluorine gas generators. During the past 15 years numerous fluorine gas generators were developed that are based on NF_4^+ salts.¹ In these systems, a highly overoxidized

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