Synthesis and characteristics of novel pentanuclear complexes $[(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2(\eta^1,\eta^5\text{-}C_5H_4CO_2)]_2M(\eta^5\text{-}C_5H_5)_2$ (M = Ti, Zr)

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The reaction of Cp_2MCl_2 complexes (M = Ti and Zr) with 2 equiv. of $(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2(\eta^5\text{-}C_5H_4COONa)$ results in the formation of the pentanuclear complexes $[(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2(\eta^1,\eta^5\text{-}C_5H_4CO_2)]_2M(\eta^5\text{-}C_5H_5)_2$, which are characterized by IR and ^1H NMR spectroscopy and cyclic voltammetry.

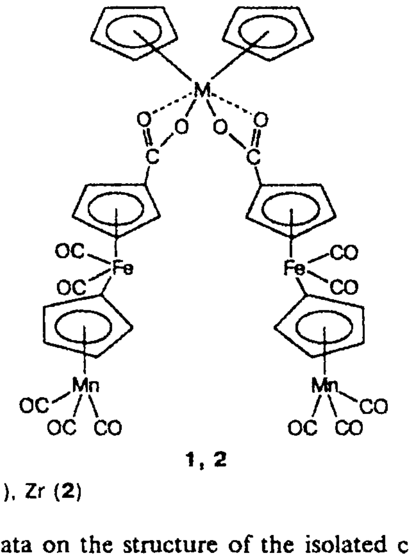
Key words: Group IV metal, polynuclear complex, synthesis; IR spectra; electrochemistry.

The synthesis and study of polynuclear complexes including diverse metals and bridged organic ligands are of special interest because of their unique conducting and catalytic properties.[1] We have previously obtained homo- and heterometallic, bi- and trinuclear transition metal complexes with bridged cyclopentadienyl ligands[2,3] linked in different ways.

In this work, the pentanuclear complexes containing metal atoms of IV, VII, and VIII Groups and bridged ligands $-OC(=O)C_5H_4$ and $\eta^1,\eta^5\text{-}C_5H_4$ were synthesized: $[(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2(\eta^1,\eta^5\text{-}C_5H_4CO_2)]_2M(\eta^5\text{-}C_5H_5)_2$, where M = Ti (**1**), Zr (**2**).

In the previous work,[4] we have shown that in the "staircase" type complexes, individual metal-containing groups mutually affect adjacent fragments resulting in the rearrangement of the electron density in the molecule. This was indicated by the frequencies of the stretching modes of the carbonyl groups in the IR spectra. It was of interest to reveal whether the regularities that were elucidated previously can be valid for the pentanuclear complexes obtained. In addition, the redistribution of the electron density should have an effect on the redox properties of these complexes; therefore, it seemed to be interesting to carry out an electrochemical study of their redox properties. The starting compound was the acid $(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2(\eta^5\text{-}C_5H_4COOH)$ that was previously synthesized[5] by the metallation of the binuclear complex $(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2Cp$ (**3**) under the action of Bu^nLi followed by the treatment with CO_2.

Complexes **1** and **2** were obtained by the reaction of Cp_2MCl_2 (M = Ti, Zr) with the sodium salt $(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2(\eta^5\text{-}C_5H_4COONa)$ (2 equiv.). They are brown and yellow powders, respectively, soluble in CH_2Cl_2, Et_2O, and benzene and insoluble in hexane.



1, 2

M = Ti (**1**), Zr (**2**)

The data on the structure of the isolated complexes and on the type of binding the fragments in the molecule were obtained from the IR spectra of the solids and solutions (Table 1). The bands were assigned to

Translated from *Izvestiya Akademii Nauk. Seriya Khimicheskaya,* No. 5, pp. 1055—1058, May, 1997.

Table 1. Frequencies of stretching modes of the CO group of the metallocarbonyl fragments and carboxylate ligand in IR spectra of complexes **1**—**3**

Complex	Solvent	ν(CO)/cm^{-1}		
		$Fe(CO)_2$	$Mn(CO)_3$	OCO
1	$CHCl_3$	2037, 2005	1985, 1923	
1	Nujol	2032, 2000	1978, 1905	1642, 1323
2	$CHCl_3$	2034, 2002	1984, 1908	
2	CH_2Cl_2	2042, 2005	1994, 1920	
2	Nujol	2035, 2004	1986, 1910	1569, 1393
3	CH_2Cl_2	2032, 2003	1978, 1916	
4	CH_2Cl_2	2012, 1960		
4	Nujol	2001, 1949, 1941		1639, 1321
5	CH_2Cl_2	2017, 1965		
5	Nujol	2017, 1995, 1971, 1939		1643, 1325, 1529, 1408

individual metallcarbonyl fragments on the basis of the regularities in the IR spectra that were discussed in the previous work.[4]

The comparison of the position of the ν(CO) bands in the IR spectra of complexes **1**—**3**, as well as the $[PhCH_2Fe(CO)_2(\eta^1,\eta^5-C_5H_4CO_2)]_2M(\eta^5-C_5H_5)_2$ complexes (M = Ti (**4**), Zr (**5**)),[6] confirms the regularities revealed: compared to ν(CO) in the spectrum of complex **3**, the frequencies assigned to the $Mn(CO)_3$ modes remain almost unchanged, while the frequencies of the $Fe(CO)_2$ fragment increase by 10—16 cm^{-1}. This testifies that the $Cp_2MOC(=O)$ fragment (M = Ti, Zr) as a whole exerts an electron-withdrawing effect on the $Fe(CO)_2$ group; however, this effect does not concern the $Mn(CO)_3$ groups. This agrees with our previous data on the trinuclear complexes[4] in which the organometallic group had no effect on the ν(CO) frequencies of remote fragments. It would be of interest to reveal the effect of the introduction of another "step" $\eta^1,\eta^5-C_5H_4Mn(CO)_3$ into the molecule on the ν(CO) values. The comparison of the frequencies of the CO stretching modes in the spectra of complexes **1** and **2** and complexes **4** and **5** [6] (see Table 1) shows that the introduction of the $\eta^1,\eta^5-C_5H_4Mn(CO)_3$ fragment into molecule instead of the benzyl substituent also increases ν(CO) of the $Fe(CO)_2$ group by 25 and 29 cm^{-1}, respectively, *i.e.*, both fragments bound to $Fe(CO)_2$ manifest electron-withdrawing properties with respect to this group.

The spectra of complexes **1** and **2** differ in the region of the vibrations of the carboxylate ligand. The spectrum of the titanium derivative **1** exhibits bands at 1642 and 1323 cm^{-1} assigned to the antisymmetric and symmetric stretching modes of the CO groups of the carboxylate ligand, respectively, while for the zirconium derivative **2**, similar vibrations are observed at 1569 and 1393 cm^{-1}. It is known[7] that the difference in the frequencies of the antisymmetric and symmetric modes of the carboxylate ligands depends on the type of coordination of these ligands: for the monodentate coordination, the difference is substantially greater (~300 cm^{-1}) than for the bidentate coordination when C—O bonds are more equilized. For complex **1**, $\Delta\nu$(as—s) is 319 cm^{-1}, which suggests the monodentate coordination of the carboxylate ligand. Previously,[6] the spectra of complex **5** contained two pairs of the bands with ν(CO) for which the $\Delta\nu$ values differed. This allowed one to conclude that one carboxylate ligand was monodentately coordinated to the Zr atom, and the coordination of the second ligand was bidentate. The IR spectrum of solid complex **2** contains only two bands corresponding to the antisymmetric and symmetric vibrations of the carboxylate groups, $\Delta\nu$(as—s) = 176 cm^{-1}, *i.e.*, it can be assumed that in this case the second O atom of the carboxylate ligand also participates in the coordination with the Zr atom (see Ref. 6: for complex **5**, $\Delta\nu$(as—s) = 121 cm^{-1}). If this difference is considered to be a measure of uniformity of the CO groups in the carboxylate ligand, the asymmetry of the carboxylate CO groups for complex **2** is greater than that for structure **5**.

The electrochemical properties of complexes **1** and **2** containing the Cp_2Ti and Cp_2Zr fragments were studied by cyclic voltammetry (CVA) using a Pt electrode in MeCN and a 0.05 *M* solution of Bu_4NBF_4 as a supporting electrolyte. These complexes are of interest because they contain atoms of three different metals: Fe, Mn, and Ti (or Zr), which can serve as electron-active centers.

The electrochemical properties of bis(cyclopentadienyl)titanium and -zirconium dichlorides (Cp_2TiCl_2 and Cp_2ZrCl_2) and the "staircase" binuclear complex **3** were studied under identical conditions to determine the sites of localization of the electronic changes during the oxidation and reduction of complexes **1** and **2**. The results obtained are presented in Table 2.

The comparison of the potentials of the first oxidation waves of complexes **1**—**3** shows that the oxidation likely occurs at the same fragment of these three molecules. We have previously studied[8] the electrochemical properties of the "staircase" complexes similar to **3** and established that the orbitals of the Mn atom participate

Table 2. Potentials of oxidation (E^{ox}) and reduction (E^{red}) peaks of the complexes studied

Complex	E^{ox}/V	E^{red}/V
1	1.10/0.97; 1.47; 1.81	0.81; 1.11; 1.72
2	1.11/0.97; 1.46; 1.76	*
3	1.08/0.96; 1.46; 1.76	1.75
$(\eta^5-C_5H_5)_2TiCl_2$	1.91	0.74; 1.70
$(\eta^5-C_5H_5)_2ZrCl_2$	2.01	1.72

Note. Conditions of electrolysis: Pt, MeCN, 0.05 *M* solution of Bu_4NBF_4, Ag/AgCl/KCl, 200 mV s^{-1}, 20 °C, *C* = 0.001 mol L^{-1}.

* No distinct reduction waves are observed.

first of all in their oxidation. This results first in the cleavage of the π-bond of the bridged ligand, and the Fe—C σ-bond cleaves only at more anodic potentials. The potentials of all three oxidation waves of complexes **1**—**3** testify that the introduction of dicyclopentadienyltitanium and -zirconium exerts almost no effect on the oxidation process, *i.e.*, according to the data of IR spectroscopy, lengthening of the polymetallic chain does not affect the change in the electron density of remote fragments.

Thus, the electrochemical oxidation of polymetallic complexes **1** and **2** occurs *via* the following scheme:

$$[(\mathrm{OC})_3\mathrm{Mn}(\eta^1,\eta^5\text{-}\mathrm{C_5H_4})\mathrm{Fe(CO)_2}(\eta^1,\eta^5\text{-}\mathrm{C_5H_4CO_2})]_2\mathrm{TiCp_2} \xrightarrow[-2\bar{e}]{\mathrm{MeCN}}$$

$$\longrightarrow [(\eta^1,\eta^5\text{-}\mathrm{C_5H_4})\mathrm{Fe(CO)_2}(\eta^1,\eta^5\text{-}\mathrm{C_5H_4CO_2})]_2\mathrm{TiCp_2} +$$

$$+\ 2\ [(\mathrm{MeCN})_2\mathrm{Mn(CO)_3}]^+$$

On the inverse scan of CVA after the oxidation peak for compounds **1**—**3**, a small cathodic peak appears, its height being ~20—30% of the anodic peak observed in the direct scanning. This can be related to the fact that the $[\mathrm{Mn(CO)_3}]^+$ cation is eliminated insufficiently rapidly, and the radical cation particles can be detected in the CVA time scale.

The cleavage of the σ-Fe—cyclopentadienyl bond occurs at the second stage:[8]

$$[(\eta^1,\eta^5\text{-}\mathrm{C_5H_4})\mathrm{Fe(CO)_2}(\eta^1,\eta^5\text{-}\mathrm{C_5H_4CO_2})]_2\mathrm{TiCp_2} \xrightarrow[-2\bar{e}]{2\ \mathrm{Cp}^{\cdot}}$$

$$\longrightarrow \{[\mathrm{Fe(CO)_2}(\eta^1,\eta^5\text{-}\mathrm{C_5H_4CO_2})]\mathrm{TiCp_2}\}^{2+}$$

The subsequent waves correspond to the further oxidation of the products formed at the second stage.

We did not study the products of the oxidation at these later stages, because the potential and site of localization of the electronic changes precisely at the first electrochemical stage, which reflects the energy of the frontier orbitals of the starting molecule and the predominant region of their localization (on the metal or ligand), are characteristic for the determination of electrochemical parameters of organometallic compounds.

The oxidation potentials of polymetallic complexes **1** and **2** and bimetallic complex **3** turned out to be very similar, while the potentials of their reduction differ substantially. For Ti-containing complex **1**, three subsequent reduction peaks are observed, and the potential of the first of them is similar to the reduction potential of $\mathrm{Cp_2TiCl_2}$. Therefore, the Ti-containing fragment is the site of localization of electronic changes in a molecule of **1**. This is indicated by the fact that complex **3** containing no Ti is reduced at much more cathodic potentials. All peaks observed during the reduction of complex **1** are chemically and electrochemically irreversible; therefore, the electron transfer to LUMO is followed by the cleavage of a bond in the complex. It is known[9,10] that depending on the nature of halogen, the radical anions $[\mathrm{Cp_2TiHal_2}]^{\cdot -}$ formed in the electrochemical reduction can decompose with both the elimination of Hal^- and detachment of the cyclopentadienyl anion. In our case, these two routes of decomposition (*a* and *b*) can be presented by the following scheme:

$$\mathrm{Cp_2TiL_2} + \bar{e} \longrightarrow [\mathrm{Cp_2TiL_2}]^{\cdot -} \begin{array}{l} \xrightarrow{a} \mathrm{CpTiL_2}^{\cdot} + \mathrm{Cp}^- \\ \xrightarrow{b} \mathrm{Cp_2TiL}^{\cdot} + \mathrm{L}^- \end{array}$$

$$\mathrm{L} = [(\mathrm{OC})_3\mathrm{Mn}(\eta^1,\eta^5\text{-}\mathrm{C_5H_4})\mathrm{Fe(CO)_2}(\eta^1,\eta^5\text{-}\mathrm{C_5H_4CO_2})]$$

In the case of route *a* on the inverse scan of CVA, the oxidation peak of the cyclopentadienyl anion should appear (−0.32 V).[11,12] However, studying the reduction of complex **1**, we observed no anodic peaks in this region, *i.e.*, in this case, route *b* is preferable.

Discussing the mechanism of the reduction of pentanuclear complex **1**, one should also consider the possibility of the process occurring at the bridged σ-bond linking the Fe atom with the cyclopentadienyl ring of the $(\eta^1,\eta^5\text{-}\mathrm{C_5H_4})\mathrm{Mn(CO)_3}$ fragment. The data of IR spectroscopy testify that the electron density on the $(\eta^1,\eta^5\text{-}\mathrm{C_5H_4})\mathrm{Fe(CO)_2}$ fragment bound to two acceptor groups $((\eta^1,\eta^5\text{-}\mathrm{C_5H_4})\mathrm{Mn(CO)_3}$ and $\mathrm{Cp_2Ti})$ is strongly lowered. This should result in facilitating the reductive cleavage of the σ-bond in $\mathrm{Fe}\text{—}\eta^1,\eta^5\text{-}\mathrm{C_5H_4Mn(CO)_3}$ compared to that in complex **3**. As can be seen from the data in Table 2, pentanuclear complex **1** is reduced at potentials almost 1 V lower than those for complex **3** in which LUMO is localized on the σ-bond of the bridge.[8] However, in our opinion, the observed difference in the reduction potentials of compounds **1** and **3** is too great, and it is difficult to explain it only by the effect of the $\mathrm{Cp_2Ti^{2+}}$ fragment on the electron density of the bridged Fe—C σ-bond. Therefore, the supposition considered above seems to be more probable: the reduction of complex **1** involves the orbitals of Ti, and the observed small difference (compared to $\mathrm{Cp_2TiCl_2}$) in the reduction potentials (see Table 2) is associated with the fact that the cyclopentadienylirondicarbonyl fragment was introduced into the molecule.

We cannot draw any conclusions on the mechanism of the reduction of the zirconium derivative **2**, because no pronounced cathodic waves were detected on the voltammogram.

Experimental

All operations were carried out in an atmosphere of argon. Hexane and $\mathrm{CH_2Cl_2}$ were dried with $\mathrm{P_2O_5}$; $\mathrm{Et_2O}$ and benzene were distilled over sodium benzophenoneketyl, $\mathrm{CDCl_3}$ was dehydrated with $\mathrm{CaCl_2}$, and $\mathrm{C_6D_6}$ was distilled over metallic Na. Solvents were distilled prior to use.

IR spectra were measured on Bruker IFS-25 and Bruker IFS-113v IR-Fourier spectrometers with a resolution of 2 cm^{-1}. ^{1}H NMR spectra were recorded on a Bruker WP-200 SY instrument. Electrochemical measurements were carried out using a PI-50 voltammetric system on a standard Pt electrode with a 0.05 *M* solution of Bu_4NBF_4 as the supporting electrolyte in anhydrous MeCN at 20 °C. Platinum was used as the accessory electrode, and a silver-chloride electrode served as the reference electrode. Oxygen was removed from a cell with a flow of dry argon. Voltammetric curves were recorded by the voltammetry method with the rates of potential scan w = 50, 100, 200, and 500 mV s^{-1}. Ohmic losses were taken into account. Acetonitrile (purity grade) was stirred over CaH_2 for 24 h and filtered, then KNO_3 (5 g) and conc. H_2SO_4 (10 mL) were added, and the resulting mixture was refluxed for 3 h and distilled. Then MeCN was refluxed above P_2O_5 for 2 h and distilled again, and the fraction with b.p. 81—82 °C (760 Torr) was collected.

Complexes 3,[13] Cp_2TiCl_2,[14] and Cp_2ZrCl_2 [15] the acid $(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2(\eta^5\text{-}C_5H_4COOH)$,[5] and the salt $(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2(\eta^1\text{-}C_5H_4COONa)$[6] were prepared by known procedures. For electrochemical measurements, complex Cp_2ZrCl_2 was twice recrystallized from benzene.

Complex $[(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2(\eta^1,\eta^5\text{-}C_5H_4CO_2)]_2TiCp_2$ (1). Cp_2TiCl_2 (0.05 g, 0.22 mmol) was added to $(OC)_3Mn(\eta^1,\eta^5\text{-}C_5H_4)Fe(CO)_2(\eta^1,\eta^5\text{-}C_5H_4COONa)$ (0.2 g, 0.47 mmol) in CH_2Cl_2 (15 mL). The reaction mixture was stirred for 24 h upon cooling with ice-cold water, then the solution was filtered off, the solvent was removed, and the residue was washed off with Et_2O. After removal of Et_2O, complex **1** (0.08 g, 40%) was isolated. Found (%): C, 48.73; H, 2.93. $C_{42}H_{26}Fe_2Mn_2O_{14}Ti$. Calculated (%): C, 49.26; H, 2.53. IR ($CHCl_3$), ν/cm^{-1}: 2035, 1985 (COFe); 2004, 1923 (COMn); 1642, 1323 (COO). ^1H NMR (C_6D_6), δ: 6.17 (s, 10 H, C_5H_5); 5.53 (t, 4 H, C_5H_4); 4.49 (t, 4 H, C_5H_4); 4.43 (t, 4 H, C_5H_4); 4.27 (t, 4 H, C_5H_4).

Complex 2 was obtained by a similar procedure using Cp_2ZrCl_2. The yield was 30%. IR ($CHCl_3$), ν/cm^{-1}: 2035, 1986 (COFe); 2004, 1920 (COMn); 1569, 1393 (COO). ^1H NMR, δ: 6.33 (s, 10 H, C_5H_5); 5.51 (m, 4 H, C_5H_4); 5.02 (m, 4 H, C_5H_4); 4.79 (m, 4 H, C_5H_4); 4.58 (m, 4 H, C_5H_4).

The content of Fe, Mn, and Ti in complex **1** and the content of Fe, Mn, and Zr in **2** were determined by the X-ray fluorescence method. The analysis was carried out on a VRA-30 spectrometer, a Mo-tube was used for excitation in a mode of 40 kV×20 mA. The crystal-analyzer was LiF (002), and the detector was (SD + FD). Analytical lines Fe-Kα; Mn-Kα; Ti-Kα — atomic ratio of metals 2 Fe : 2 Mn : Ti. Analytical lines Fe-Kα; Mn-Kα; Zr-Kα — atomic ratio of metals 2 Fe : 2 Mn : Zr.

This work was financially supported by the Russian Foundation for Basic Research (Project Nos. 96-03-34082 and 94-03-08628a).

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Received December 27, 1996;
in revised form January 30, 1997