SILVER AND COPPER SALTS OF  $\alpha$ ,  $\beta$ -UNSATURATED PERFLUOROCARBOXYLIC ACIDS IN DECARBOXYLATION REACTIONS

V. F. Cherstkov, M. V. Galakhov, E. I. Mysov, S. R. Sterlin, and L. S. German UDC 542.97:547.464.6-38'161

5. R. Sterrin, and L. S. German

The pyrolysis of Ag perfluorocarboxylates is known to result in the formation of perfluoroalkanes, which are products of the dimerization of radicals that are formed by oxidative decarboxylation of the initial salts [1, 2]. The extension of this reaction to salts of  $\alpha$ , $\beta$ -unsaturated perfluorocarboxylic acids opens up a new route for the synthesis of perfluorinated conjugated dienes.

It transpired that thermal decomposition of Ag salts of perfluoro-4-methylpent- (Ia) and perfluoro-4,4-dimethylpent-2-enoic (Ib) acids resulted in the formation of the perfluorodienes (IIa-b) in 87-89% yield

 $\begin{array}{c} R_{\rm F} CF = CFCOOAg \xrightarrow{210-220^{\circ}} R_{\rm F} CF = CF - CF = CFR_{\rm F} \\ (Ia-b) & (IIa-b) \\ R_{\rm F} = i - C_3 F_7 \ (a); \ t - C_4 F_9 \ (b). \end{array}$ 

According to the data of <sup>19</sup>F NMR, diene (IIa) is 87% and diene (IIb) is completely in the form of the (1E,3E)-S-cis isomer, as follows from the equal magnitude of the <sup>19</sup>F spin-spin coupling constants (Table 1) relative to the corresponding values in 1,4di(p-anisyl)tetrafluorobutadiene [3], which also exists in the form of its (1E,3E)-S-cis isomer [4]. These compounds were also obtained by decarboxylation of the analogous copper carboxylates (IIIa, b), but the process is less selective and the composition of the reaction products depends on the conditions for decarboxylation.\* Thus, in the absence of a solvent, on thermal composition of the copper salt of perfluoro-4-methylpent-2-enoic acid (IIIa), perfluoro-4-methylpent-2-enoyl fluoride is formed together with diene (IIa) and unidentified high-boiling products. The yield of the diene under these conditions is not greater than 10%. However, in high-boiling organic solvents such as N-methylpyrrolidone (MP) or sulfolane, thermolysis of (IIIa) occurs quite smoothly and diene (IIa) is formed (virtually pure (1E,3E)-S-cis isomer) (isomer A) in 45% yield. The only by-product of the reaction in this case is 1-hydro-3-trifluoromethylperfluoro-1-butene (IVa). Evidently, decarboxylation of carboxylate (IIIa) occurs in two stages. At the first, one of the two carboxylate groups is decarboxylated to give diene (IIa) and the Cu<sup>+</sup> carboxylate, which is then decarboxylated to give the organocopper derivative (V). Reaction of the latter with the solvent gives hydride (IVa). A mixture of diene (IIb) and 1-hydro-3,3-bis(trifluoromethyl)perfluoro-l-butene (IVb) is obtained in a similar manner by decomposition of  $Cu^{2+}$  perfluoro-4,4-dimethylpent-2-enoate in MP

 $\begin{array}{c} (\mathrm{R}_{\mathrm{F}}\mathrm{CF}{=}\mathrm{CF}\mathrm{COO})_{2}\mathrm{Cu} \xrightarrow{160-165^{\circ}}_{\mathrm{MP}; -\mathrm{CO}_{2}} [\mathrm{R}_{\mathrm{F}}\mathrm{CF}{=}\mathrm{CF}\mathrm{COOCu} + \mathrm{R}_{\mathrm{F}}\mathrm{CF}{=}\mathrm{CF}^{\circ}] \\ (\mathrm{IIIa, b}) & \Delta \downarrow {-}\mathrm{Co}_{2} & \downarrow \\ \mathrm{R}_{\mathrm{F}}\mathrm{CF}{=}\mathrm{CF}\mathrm{H} \xrightarrow{\mathrm{MP}}_{\mathrm{F}} [\mathrm{R}_{\mathrm{F}}\mathrm{CF}{=}\mathrm{CF}\mathrm{Cu}] & (\mathrm{IIa, b}) \\ (\mathrm{IVa, b}) \\ \mathrm{R}_{\mathrm{F}} = i{-}\mathrm{C}_{3}\mathrm{F}_{7} \text{ (a), } i{-}\mathrm{C}_{4}\mathrm{F}_{8} \text{ (b).} \end{array}$ 

<sup>\*</sup>In thermal decarboxylation reactions, salts (IIIa, b) resemble  $Cu^+$  and  $Cu^{2+}$  salts of aromatic acids, which decompose to form the corresponding biaryls in 8-60% yield [5, 6]. Stilbene was obtained by decomposition of  $Cu^{2+}$  2,3-diphenylacrylate [7]. The main product of the decarboxylation of  $(C_3F_7CO_2)_2Cu$  was pentafluoropropionyl fluoride; the formation of perfluorohexane was not recorded [2].

A. N. Nesmeyanov Institute of Heteroorganic Compounds, Academy of Sciences of the USSR, Moscow. Translated from Izvestiya Akademii Nauk SSSR, Seriya Khimicheskaya, No. 6, pp. 1336-1340, June, 1989. Original article submitted March 30, 1988.

		စ်	mqq		Spin	-spin	coupl	ing co	nstant,	Hz	
Compound		2	e	4	1-2	1-3	1-4	23	2-4	34	J, Hz
CF <sub>3</sub> <sup>1</sup>	-0,2	111,6	77,4	77,2	7,5	7,5	4,0	+6,6	-41,3	051-	$J_{i-i} = -33,0$
$CF_3$ $CF_3$ $F^4$ $F^{3\prime}$ $CF_3^{1\prime}$ (IIa)				-				<u> </u>			$J_{3-4} = +12,5$
4.F CF2 CF3											
$(CF_3^1)_3C \qquad F^3 \qquad (11b)$	-10,0	$\Delta v = 11$	75 0, Hz		ก	4	7	- 137,2	, 	1	$J_{3-3}^{3-3} = -26,8$ $J_{2-2}^{2} = +41,3$ $J_{2-3}^{2} = +8,9$
$\mathbf{F}^{2}$ $\mathbf{r}$ $\mathbf{F}$ $\mathbf{C}(\mathbf{CF}_{3}^{T})_{3}$	43 ½	6	Co		6.61	л Ц	)	135			75
$CF_{3^{1}} \bigvee_{C} \bigvee_{C^{3}} (IV b)$	тю́Г-	<b>76</b> .	00	1	7,21	1,01		507			$J_{F^2-H} = 13$
GF3 C						******					
$(CF_3^1)_2 CF^2 \xrightarrow{F^4} (VII)$	-0,3	110,0	81,3	32,2	7,5	9,4	4,7	13,1	44,2	132,5	
aF Cl (CF <sub>3</sub> ) <sub>2</sub> CF F*	6.1-	107,5	89,9	6()9	7,5	8,5	6,5	10,5	46,0	130	

\*Spectrum obtained with <sup>19</sup>F-<sup>1</sup>H.

 $^{19}F$  NMR Spectra of Fluoroolefins (CF<sub>3</sub>COOH used as external standard) TABLE 1.

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The formation of the intermediate organocopper derivative (V) is confirmed by the formation of 1-phenyl-3-trifluoromethylperfluoro-1-butene (VI) by decarboxylation of carboxylate (IIIa) in MP in the presence of iodobenzene\*

$$(IIIa) + PhI \xrightarrow{105-125^{\circ}, MP} PhCF = CFCF(CF_3)_2 + (IIa) + (IVa)$$

$$(VI)$$

According to the proposed scheme for the decomposition of carboxylates (III), perfluorodienes (II) are formed as a result of the oxidative decarboxylation of  $Cu^{2+}$  salts, and hence their yield cannot exceed 50% of the initial carboxylic acid. It was to be expected that the yield of dienes (II) would increase if the decomposition of the carboxylates were to be carried out in the presence of inorganic  $Cu^{2+}$  salts such as halides, which by undergoing an exchange reaction with the initial salts (III) or by oxidizing the univalent copper carboxylates that are formed at the first stage of decarboxylation would give asymmetric  $Cu^{2+}$  haloperfluoroalkenoates. Decomposition of the latter should mainly lead to products that are derivatives of the corresponding perfluorovinyl radicals [1-chloroolefins and dienes (II)] as a result of a decrease in the yield of hydrides (IV).

We studied the thermal decarboxylation of a mixture of (IIIa) and  $CuCl_2$  in MP. In fact, the relative content of hydride (IVa) in the reaction products decreased sharply (14% yield) but the main reaction product was not diene (IIa) (yield 23%) but 1-chloro-3-trifluoromethylperfluoro-1-butene (VII) (yield 36%). The decrease in yield of hydride (IVa) indicates that the carboxylate groups mainly formed part of salts (IIIa) or (VIII). Oxidative decarboxylation of these salts leads to the formation of perfluoroisopentenyl radicals (IX), which are mainly destroyed not by coupling as in the case of the thermolysis of pure carboxylate (IIIa) but by removal of a Cl atom from CuCl<sub>2</sub> or salt (VIII)

$$(111a) \xrightarrow{\text{CuCl}_2} [(CF_3)_2 CFCF = CFCOOCuCl] \xrightarrow{\Delta} [(CF_3)_2 CFCF = CF'] \rightarrow (11a)$$

$$\Delta \downarrow \stackrel{-CO_2}{-(1X)} \quad \text{CuCl} \downarrow \stackrel{(VIII)}{\text{CuCl}_2} \xrightarrow{-CuCl} \qquad \downarrow \stackrel{CuCl_2}{\text{Or}} \xrightarrow{(VIII)} [(CF_3)_2 CFCF = CFCOOCu] \xrightarrow{\Delta} [Va] \xrightarrow{MP} (1Va) \qquad (CF_3)_2 CFCF = CFCI \qquad (VII)$$

## EXPERIMENTAL

 $^{19}{\rm F}$  NMR spectra were recorded on Bruker CXP-200 and WP-200SY spectrometers with operating frequency 188.3 MHz. Homonuclear suppression was carried out with time distribution using a B-SV3BX spin decoupling generator (pulsed suppression power 10 W). Spectra of compounds (IIa, b) were calculated using the standard program PANIC on an Aspect-2000 computer to a root-mean-square iteration error value of RMS = 0.1 Hz using the relative signs of the spin-spin coupling constants of perfluorobutadiene [9]. Mass spectra were obtained on a VG-7070E chromatograph-mass spectrometer (electron energy 70 eV; source temperature 160°C; OV-101 capillary column; length 25 m). Raman spectra were recorded on a Ramanor-1000 spectrometer.

<u>Preparation of Salts (Ia, b) and (IIIa, b)</u>. To an aqueous solution of the Na salt of perfluoro-4-methylpent-2-enoic acid or perfluoro-4,4-dimethylpent-2-enoic acid [10] was added an aqueous solution of  $AgNO_3$  (5-8 mole % excess). The precipitate that formed was extracted with ether, the extract was dried, and the salt was dried in a vacuum desiccator over concentrated H<sub>2</sub>SO<sub>4</sub>. In a similar manner, when aqueous solutions of Na salts of unsaturated perfluorocarboxylic acids were treated with an excess of a saturated aqueous solution of CuSO<sub>4</sub> followed by extraction with ether, salts (IIIa-b) were obtained, and they were dried in a vacuum-drying Fisher apparatus at the boiling point of CCl<sub>4</sub>.

Decomposition of Salts (Ia, b). Salt (Ia) (10.4 g, 20 mmoles) was heated with the flame of a gas jet (decomposition temperature 210-220°C) and the pyrolysis products were collected in a cooled receiver. The pyrolyzate was washed with dilute  $HNO_3$  and dried over MgSO<sub>4</sub>. Yield was 6 g of a mixture which, according to the GLC data, contained diene (IIa) (91%), hydride (IVa) (4%) (identified by comparison with a known sample [11]), and unidentified reaction products (5%), from which (IIa) was isolated by fractionation, bp 124-126°C. Found, %: C 25.86; F 74.16.  $C_{10}F_{18}$ . Calculated, %: C 25.97; F 74.02. Raman spectrum

 $<sup>*</sup>PhC_6H_4NO_2-2$  was obtained in 39% yield by decarboxylation of  $2-O_2NC_6H_4CO_2Cu$  in the presence of iodobenzene [8].

 $(CC1_4): v_{max} = 1716 \text{ cm}^{-1} (C=C). According to the data of GLC and chromatography-mass spectrometry, (IIa) was a mixture containing 87% of isomer (A) and 13% of a difficult-to-separate mixture of two other isomers, whose structure was not precisely established. The mass spectra of all the compounds were identical. Mass spectrum of isomer A, m/z (rel. intensity, %): 462 M<sup>+</sup> (100), 443 [M<sup>+</sup> - F] (13.6), 424 [C_9F_{16}]<sup>+</sup> (1.9), 393 [C_9F_{15}]<sup>+</sup> (96.3), 373 [C_9F_{14}]<sup>+</sup> (2.2), 355 [C_9F_{13}]<sup>+</sup> (17.9), 343 [C_8F_{13}]<sup>+</sup> (3.6), 312 [C_7F_{12}]<sup>+</sup> (7.4), 305 [C_8F_{11}]<sup>+</sup> (19.2), 293 [C_7F_{11}]<sup>+</sup> (100), 286 [C_8F_{10}]<sup>+</sup> (2.3), 274 [C_7F_{10}]<sup>+</sup> (2.7), 267 [C_8F_9]<sup>+</sup> (5.9), 255 [C_7F_9]<sup>+</sup> (30.4), 243 [C_6F_9]<sup>+</sup> (100), 235 [C_7F_8]<sup>+</sup> (16.5), 224 [C_6F_8]<sup>+</sup> (11), 219 [C_4F_9]<sup>+</sup> (2.3), 217 [C_7F_7]<sup>+</sup> (15.1), 212 [C_9F_8]<sup>+</sup> (1.1), 205 [C_6F_7]<sup>+</sup> (14.8), 198 [C_7F_6]<sup>+</sup> (2.1), 193 [C_5F_7]<sup>+</sup> (14.3), 186 [C_6F_6]<sup>+</sup> (19.5), 181 [C_4F_7]<sup>+</sup> (11.5), 179 [C_7F_5]<sup>+</sup> (3.3), 174 [C_5F_6]<sup>+</sup> (2.6), 169 [C_3F_7]<sup>+</sup> (1.1), 167 [C_6F_5]<sup>+</sup> (8.5), 162 [C_4F_6]<sup>+</sup> (2.1), 155 [C_5F_5]<sup>+</sup> (27.6), 148 [C_6F_4]<sup>+</sup> (4.5), 143 [C_4F_5]<sup>+</sup> (11.5), 136 [C_5F_4]<sup>+</sup> (1.9), 131 [C_3F_5]<sup>+</sup> (30.9), 129 [C_6F_3]<sup>+</sup> (2.4), 100 [C_2F_4]<sup>+</sup> (100), 98 [C_5F_2]<sup>+</sup> (11.5), 117 [C_5F_3]<sup>+</sup> (17.1), 112 [C_3F_4]<sup>+</sup> (3.3), 105 [C_4F_3]<sup>+</sup> (2.4), 100 [C_2F_4]<sup>+</sup> (100), 98 [C_5F_2]<sup>+</sup> (1.8), 93 [C_3F_3]<sup>+</sup> (22.7), 86 [C_4F_2]<sup>+</sup> (1.3), 74 [C_3F_2]<sup>+</sup> (3.1), 69 [CF_3]<sup>+</sup> (100), 50 [CF_2]<sup>+</sup> (3.4). In a similar manner decomposition of 12 g (27 mmoles) of salt (Ib) gave 5.7 g (867) of (IIb), mp 89-90°C (hexane). Found, %: C 26.16; F 74.19. C_{12}F_{22}. Calculated, %: C 25.63; F 74.37. Raman spectrum (CC1_4): v_{max} = 1706 cm<sup>-1</sup> (C=C).$ 

Decomposition of Salts (IIIa, b). A solution of 16.4 g (26 mmoles) of salt (IIIa) in 30 ml of anhydrous MP was agitated at 160-165°C until evolution of gas had stopped and the reaction products had distilled off. The pyrolyzate was washed with dilute HCl and water and dried over MgSO<sub>4</sub>. Hydride (IVa) (5 g, 42%) was separated by distillation, bp 37-41°C, as was diene (IIa) (5.8 g, 47%), bp 123-127°C. In a similar manner, decomposition of 6 g (8.4 mmoles) of salt (IIIb) in 15 ml MP at 140-190°C gave 1.2 g (25%) of hydride (IVb) (identified by comparison with a known sample using GLC) and 2.2 g (46%) of diene (IIb). Hydride (IVb) was obtained according to the method of [9] by decomposition of the Na salt of perfluoro-4,4-dimethylpent-2-enoic acid in diethylene glycol at 160-180°C, yield 79%, bp 64-65°C. Found, %: C 25.46; H 0.32; F 73.97. C<sub>6</sub>HF<sub>11</sub>. Calculated, %: C 25.52; H 0.35; F 74.11. IR spectrum ( $\nu_{max}$ , cm<sup>-1</sup>): 1710 (C=C); 3125 (C-H).

Decomposition of Salt (IIIa) in the Presence of Iodobenzene. A mixture of 12.28 g (20 mmoles) of salt (IIIa), 8.2 g (40 mmoles) of iodobenzene, and 50 ml of anhydrous MP was agitated for 6 h at 105-112°C, then for 3 h at 120-125°C. The products that were volatile at 150°C (100 mm) were distilled off into a receiver and trap (-78°C), the contents of the receiver and trap were combined, the lower layer was separated, and 3.35 g of diene (IIa) was obtained. The upper layer was washed with dilute HC1, extracted with  $CH_2Cl_2$ , and the extract was dried over MgSO<sub>4</sub> and distilled. A mixture (2.3 g) was obtained, which contained, according to GLC, an olefin (VI) (47%), iodobenzene (50%), and diene (IIa) (3%). The mixture was distilled twice over concentrated  $H_2SO_4$  and the fraction with bp 171-172°C was isolated, containing 95% (VI), 4% iodobenzene, and 1% diene (IIa). IR spectrum:  $v_{max} = 1695$  cm<sup>-1</sup> (C=C). Mass spectrum, m/z (rel. intensity, %): 308 M<sup>+</sup> (100), 289 [M<sup>+</sup> - F] (8.4), 239 [M<sup>+</sup> - CF<sub>3</sub>] (91.3), 220 [C<sub>10</sub>H<sub>5</sub>F<sub>5</sub>]<sup>+</sup> (8.4), 219 [C<sub>10</sub>H<sub>4</sub>F<sub>5</sub>]<sup>+</sup> (65.6), 200 [C<sub>4</sub>F<sub>8</sub>]<sup>+</sup> (18.1), 189 [C<sub>9</sub>H<sub>5</sub>F<sub>4</sub>]<sup>+</sup> (5.5), 170 [C<sub>9</sub>H<sub>5</sub>F<sub>3</sub>]<sup>+</sup> (18.5), 169 [C<sub>9</sub>H<sub>4</sub>F<sub>3</sub>]<sup>+</sup> and [C<sub>3</sub>H<sub>7</sub>]<sup>+</sup> (75.1), 151 [C<sub>9</sub>H<sub>5</sub>F<sub>2</sub>]<sup>+</sup> (17.1), 105 [C<sub>4</sub>F<sub>3</sub>]<sup>+</sup> (14.1), 77 [C<sub>6</sub>F<sub>5</sub>]<sup>+</sup> (18.4), 69 [CF<sub>3</sub>]<sup>+</sup> (19.5).

Decomposition of Salt (IIIa) in the Presence of CuCl<sub>2</sub>. A mixture of 12.2 g (20 mmoles) of salt (IIIa), 4.85 g (36 mmoles) of CuCl<sub>2</sub>, and 50 ml of anhydrous MP was agitated at 145-175°C and the light-boiling reaction products were distilled off at the same time. A mixture (7.2 g) was obtained, which according to GLC contained hydride IVa (29%), a chloroolefin (VII) (52%), and a dimer (IIa) (18%). The yields of (IVa), (VII), and (IIa) were 14, 36, and 22.5%, respectively. Fractionation of the mixture gave (VII), bp 59-61°C. Found, %: C 22.59; F 64.17.  $C_5$ ClF<sub>9</sub>. Calculated, %: C 22.51; F 64.17.

## CONCLUSIONS

Thermal decarboxylation of the Ag and  $Cu^{2+}$  salts of  $\alpha,\beta$ -unsaturated perfluorocarboxylic acids results in the formation of perfluoro-1,3-dienes, which are products derived from the dimerization of intermediate vinyl radicals.

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## REACTIONS OF BORYLOXYMETHYL- AND HYDROXYMETHYLPHOSPHINES

WITH AMINES

G. N. Nikonov, A. S. Balueva, O. A. Erastov,\* UDC 542.91:547.1'127'118:547.89 and B. A. Arbuzov

Previously it was shown that boryloxymethylphosphines react with primary amines with the formation of aminomethylphosphines. In the reaction of aromatic amines and benzylamine with 2,5-diphenyl-1,3,2,5-dioxaboraphosphorinane, 5-(hydroxymethyl)-2,2,5-triphenyl-1,3,2,5dioxaborataphosphoniarinane, and ammonium 2,2,5-triphenyl-1,3,2,5-dioxaborataphosphorinane, the corresponding 1,3,5-diazaphosphorinanes, 1,5,3,7-diazadiphosphacyclooctanes, and bis-(aminomethyl)phosphines are obtained, depending on the reagent ratios [1-3]. The reaction of primary amines and bis(hydroxymethyl) phenylphosphine affords the same compounds [4]. No differences were observed in the reactivity of boroxymethyl- and hydroxymethylphosphines. Even in the case of 5-(hydroxymethyl)-1,3,2,5-dioxaborataphosphoniarinane, where the fourcoordinate boron atom stabilizes the molecule, it was not possible to carry out the reaction only with participation of the hydroxymethyl group.

The number of amines used in [1-4] is rather limited; therefore, we could expect that its expansion with weakly basic amines would make it possible to elucidate the characteristics of the reactions of boryloxymethyl- and hydroxymethylphosphines. For this purpose, we studied the reactions of boryloxy- and hydroxymethylphosphines with diphenylamine and o-, m-, and p-aminobenzoic acids.

In the reaction of diphenylamine with 5-(hydroxymethyl)-2,2,5-triphenyl-1,3,2,5-dioxaborataphosphoniarinane (I) with reagent ratio 1:1, we recovered a crystalline product not having absorption of hydroxyl groups in the IR spectra and with chemical shift of the P-atom signal in the <sup>31</sup>P NMR spectrum equal to 5 ppm. The data of elemental analysis indicated that the molecule contained nitrogen atoms and corresponded to 5-(diphenylaminomethyl)-2,2,5triphenyl-1,3,2,5-dioxaborataphosphoniarinane (II)



<sup>\*</sup>Deceased.

A. E. Arbuzov Institute of Organic and Physical Chemistry, Kazan Branch, Academy of Sciences of the USSR. Translated from Izvestiya Akademii Nauk SSSR, Seriya Khimicheskaya, No. 6, pp. 1340-1343, June, 1989. Original article submitted December 4, 1987.