OsO_3F_2 and OsO_2F_4, Preparation and Crystal Structures

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A new method for preparing OsO_3F_2 is presented. Its crystal structure reveals a polymeric chain structure with distorted octahedral geometry for the OsO_3F_3 units and symmetrical, non-linear fluorine bridges. The OsO_3F_3 unit has a facial geometry. This structure is not in accord with predictions on the molecular structure of OsO_3F_2. OsO_2F_4, obtained from KrF_2 and OsO_4, crystallizes in the space group $P6_1$ forming a sixfold helical chain arrangement of nearly octahedral OsO_2F_4 molecules. Oxygen or fluorine atoms are partially disordered. The *cis* orientation of the two oxygen atoms is established by means of NMR, vibrational spectroscopy, and X-ray data.

The valence shell electron pair repulsion (VSEPR) model describes successfully the geometry of main group molecules, cations and anions, if the coordination number six is not exceeded. In recent years it has become obvious that this model sometimes fails with d^0 transition compounds. The most striking examples are the trigonal prismatic structures of $W(CH_3)_6$ and $Zr(CH_3)_6^{2-}$[1,2]. The reason for this remarkable deviation is at present not fully understood. Another severe case where the VSEPR model fails to predict the correct geometry is the *cis* orientation of the oxygen atoms in $AO_2X_4^{n-}$ anions, like $MoO_2F_4^{2-}$[3], $WO_2F_4^{2-}$[4,5], and $ReO_2F_4^{-}$[6]. One of the subjects of the present investigation is to check osmium(VIII) fluoride oxide structures relative to this structural model, because structures with small or better even zero charge on the molecular unit are least influenced by lattice energy. The highest oxidation state VIII is only reached by Os, Ru, and Xe, of which Os(VIII) compounds are by far the most stable ones. But in spite of this there is a surprising paucity of structural data of Os(VIII) compounds. OsO_3F_2 is known since 1957[7], while OsO_2F_4 has only very recently been obtained and was first incorrectly identified as $OsOF_6$[8,9]. In the course of this investigation we learned that there were ab initio calculations on OsO_4, OsO_3F_2, OsO_2F_4, $OsOF_6$, and OsF_8 in progress[10]. These results are presented in the preceding paper and showed that $OsOF_6$ and OsF_8 are unlikely to exist[10].

OsO_3F_2

OsO_3F_2, which was first described by Hepworth and Robinson, was prepared by fluorination of OsO_4 with BrF_3[7].

The existence of the compound has been clearly established by elemental analysis, IR, Raman and mass spectroscopy[7,11,12]. However, the solid-state structure remained unclear. From X-ray powder data three modifications of OsO_3F_2 have been established[13]. The high-temperature orthorhombic phase might be similar to that of $MoOF_4$[14,15], the intermediate-temperature orthorhombic phase might be similar to RuF_5[16]. The crystal structure of the low-temperature monoclinic phase is described in this paper.

The low vapor pressure of OsO_3F_2 is an indication for a bridged dimer or polymer structure in the solid, if compared with the vapor pressures of OsO_4 and OsO_2F_4. The VSEPR model predicts a trigonal-bipyramidal structure for the OsO_3F_2 molecule in the gas phase with all oxygens positioned in the equatorial plane (see Figure 1). In fact, matrix-isolated OsO_3F_2 has been assigned such a D_{3h} structure, based on Raman and IR measurements[11,12]. Also, ab initio calculation predicts this structure for the molecule[10]. The very elusive XeO_3F_2 is thought indeed to have a trigonal-bipyramidal structure with all oxygens positioned equatorially according to vibrational studies, but its instability prevented further structural confirmations[17,18].

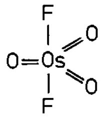

Figure 1. Proposed structure for the OsO_3F_2 gas molecule

For the present investigation OsO_3F_2 was prepared by reaction of an excess of liquid ClF_3 with OsO_4: Osmium pentafluoride oxide, $OsOF_5$, which was also formed, was separated by sublimation. Crystals of OsO_3F_2 were grown by sublimation, and the crystal structure was determined by the X-ray method. The structural determination was straightforward if the extreme absorption was corrected. The quality of the structure was good enough to clearly distinguish oxygen from fluorine atoms, mainly by the much shorter bond lengths of Os–O versus Os–F bonds. Also the

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fact that an exchange of oxygen for fluorine atoms or vice versa results in too small and too large vibrational amplitudes as well as larger *R* values confirm the oxygen and fluorine assignments. The crystal structure contains a zigzag chain of OsO_3F_3 units that are interconnected by almost symmetrical bent *cis* fluorine bridges (see Figure 2). The oxygen and fluorine triplets have a facial (*all-cis*) orientation, with O—Os—O bond angles considerably larger than 90° (102–104°) and F—Os—F bond angles smaller than 90° (77°). The electron pair repulsion model predicts a meridial geometry for an OsO_3F_3 unit. But in spite of the fact that this model fails to predict the overall structure correctly, it is nevertheless successful in predicting the larger than 90° O—Os—O and smaller than 90° F—Os—F angles, based on the assumption of the larger electron density in the OsO bond versus the OsF bond. It should be noted that the observed solid-state structure is in accord with the vibrational spectra of solid OsO_3F_3[19], and that a very recent OsL_{III} EXAFS study reproduced the Os=O and Os–F bond lengths quite accurately[20].

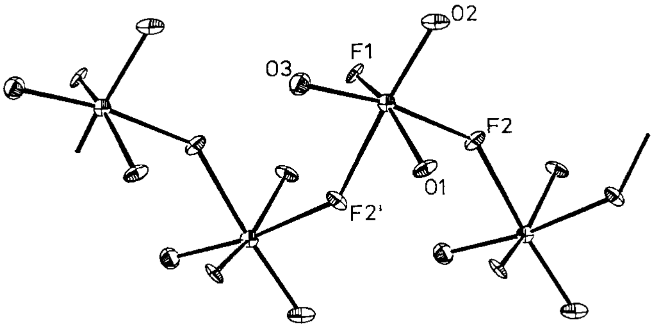

Figure 2. Chain structure of $(OsO_3F_2)_x$, ORTEP plot, ellipsoids show 50% probability. Selected bond lengths [pm]: Os–O(1,2,3) 172.7(1), 168.8(1), 167.8(1), Os–F(1,2,2'), 187.9(1) 212.6(1), 210.8(1); selected bond angles [°] O=Os=O (1–2, 1–3, 2–3) 104.2(5), 101.3(5), 102.0(5), F–Os–F(1–2, 1–2', 2–2') 77.4(4), 76.4(3), 79.6(3), Os–F(2)–Os' 143.9(2)

This solid-state structure of OsO_3F_2 shows again that the coordination number five is less favored than CN six (or four). It also shows again that bridging among transition metal oxyfluorides occurs in general by fluorine bridges. This has already been noted for the structures of WOF_4, $MoOF_4$, $ReOF_4$, $TcOF_4$[14,15] and is in sharp contrast to main group oxyfluorides which inevitably show oxygen bridges, e.g. $(SeOF_4)_2$, $(TeOF_4)_2$, $(IO_2F_3)_2$[21–23]. This may result from the substantially larger bond energy of transition metal-oxygen double bonds versus heavy main group-oxygen double bonds when compared with the respective single bonds. The facial (*all-cis*) arrangement of oxygen and fluorine atoms in OsO_3F_2 comes as a surprise, also because the postulated D_{3h} structure of gaseous OsO_3F_2 cannot be considered a cut-out of the solid-state structure. If the D_{3h} gas structure proves to be correct, then a severe reorientation of ligands is necessary upon sublimation. In this connection it should be noted that gaseous OsO_3F_2 has a static electric dipole moment, the only indication so far that the predicted D_{3h} structure might not be correct[24].

The preference of *cis* oxygen and *cis* fluorine positioning can be explained by a *trans* effect. This means that fluorine and doubly bonded oxygen atoms should be *trans* to each other, because only in this situation each double bond to oxygen will not have to share a d-orbital with another oxygen double bond. Such a bonding scheme would call for a lenghtening of the *trans*-to-oxygen positioned metal-fluorine bonds as compared to *trans*-to-fluorine metal-fluorine bonds. In the case of OsO_3F_2, however, there are only *trans*-to-oxygen metal fluorine bonds, and their remarkable bond length difference is clearly a result of participation and non-participation in bridge formation.

OsO_3F_2 has been treated with KF, RbF, or CsF to give the $OsO_3F_3^-$ anion[7,25]. The analysis of the vibrational spectra favors a facial geometry[26,27] that would indeed be expected in view of the OsO_3F_2 structure. However, isoelectronic $ReO_3F_3^{2-}$ is assigned a meridial geometry, according to its vibrational spectra[28].

OsO_2F_4

This compound was only recently prepared for the first time[8,9] after it was at first incorrectly identified as $OsOF_6$[8]. The preparation needs KrF_2 as the fluorinating agent:

$$OsO_4 + 2\ KrF_2 \xrightarrow{HF} OsO_2F_4 + 2\ Kr + O_2$$

The error was due to a lack of NMR data and the misinterpretation of the elemental analysis of the new compound. It is possible that OsO_2F_4 obtained from HF solutions contains two HF molecules, so the analytical Os/F ratio is 1:6. This seems to be a classical mistake: The reported compounds CrF_6[29–37], OsF_7[38], OsF_8[39,40] and XeF_8[41,42] may have been in fact CrF_5, OsF_6, and XeF_6 containing HF impurities.

The A_2B_2-type ^{19}F-NMR spectrum of OsO_2F_4 clearly indicates a *cis* oxygen position (Figure 3). This prediction can be confirmed by the vibrational spectra: The occurrence of the same bands in the IR and Raman spectra and also the presence of obviously two Os=O stretching vibrations at 942 and 932 cm^{-1} definitely rule out a D_{4h} (*trans*) symmetry. The *cis* geometry is also predicted by a recent ab initio calculation[10].

In the light of the known trigonal-pyramidal structures of the d^0 compounds $W(CH_3)_6$ and $Zr(CH_3)_6^{2-}$ [1,2] an octahedral structure was not completely certain. Obviously, such deviations from the octahedral structure can occur if the bond polarity is quite small. Another candidate for such behavior might be CrF_6, if it exists[31,35], although very recent calculations favor octahydral symmetry[36,37].

The crystal structure of OsO_2F_4, however, reveals that the octahedral geometry is indeed retained. It was expected that the crystallinity of a molecule with six almost equal ligands and only weak intermolecular interactions would be poor. Most known octahedral hexafluorides crystallize in a plastic cubic high-temperature phase and in a partly ordered orthorhombic low-temperature phase[43]. The related compound $OsOF_5$ shows the same behavior, and in the low-

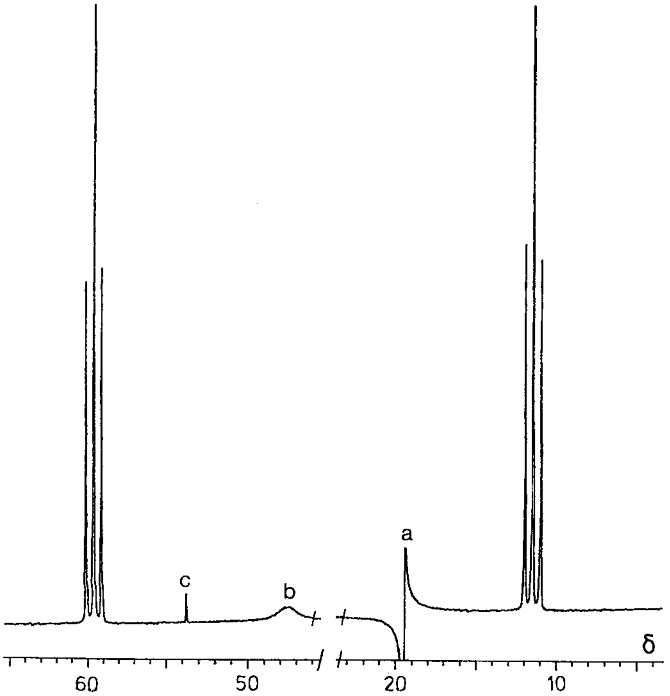

Figure 3. ^{19}F-NMR spectrum of OsO_2F_4 in HF solution. The occurrence of two triplets clearly establishes a *cis* geometry. – a: Folding of the HF signal; b: KrF_2; c: OsO_3F_2

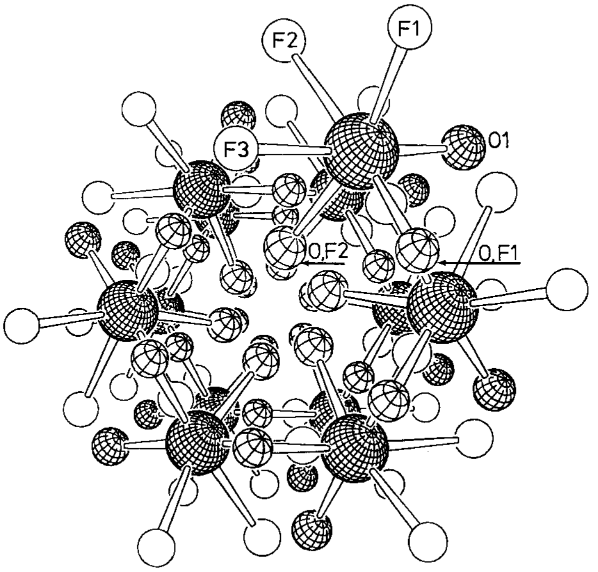

Figure 4. View down the hexagonal *c* axis of OsO_2F_4, XP plot. Osmium and oxygen atoms are shaded, the disordered oxygen/fluorine atom pair is partly shaded

temperature phase the oxygen atom is disordered in two positions[44].

It was surprising, that OsO_2F_4 crystallizes in a completely different system. Crystals are hexagonal of space group $P6_1$ or $P6_5$. These space groups enforce a right- or left-handed helical chain orientation of the molecules with six molecules in the repeating unit (Figure 4).

The X-ray structure determination was severely hampered both by problems of absorption, due to the fact that single crystals grew only as thin hexagonal platelets, and by disorder. Probably because of errors caused by absorption that in spite of a correction could not be completely accounted for, it was not possible to differentiate between space groups $P6_1$ and $P6_5$. Also the calculation as a meroedric twin $P6_1/P6_5$ did not give better or worse results than those presented below. The result, however, allowed us to eliminate space groups $P6_122$ or $P6_522$ as possible alternatives, for which to our knowledge at least one example among pure inorganic compounds is known: AuF_3 and possibly AgF_3[45,46].

The helical chain is composed of OsO_2F_4 octahedra (Figure 5). There are four noticeable intermolecular contacts between ligand atoms of 273.9, 271.3, 276.1, and 276.3 pm length. They are only slighly longer than the intramolecular contacts between ligand atoms (256.5 – 271.9 pm). Because all ligand atoms have these intermolecular contacts it is not possible to distinguish between oxygen and fluorine in this way. The assignment of the ligand atoms as shown in Figures 4 and 5 is based, however, on intramolecular bond lengths and bond angles. There are one short, 164.2(25), two intermediate, 180.5(23), 182.4(23), and three long bonds 186.7(22), 194.2(16), 195.2(36) pm. We have assigned the atom with the short bond length to an oxygen atom, the two atoms with intermediate bond length to a disordered O,F pair, and the rest to fluorine atoms. From this assignment result two possible pairs of oxygen atoms that both are *cis*-oriented, as demanded by the ^{19}F-NMR spectra. These pairs also possess the large ligand-osmium-ligand angles of 96.1 and 95.1° within the octahedron, as is required by the electron pair repulsion model and the ab initio calculation[10].

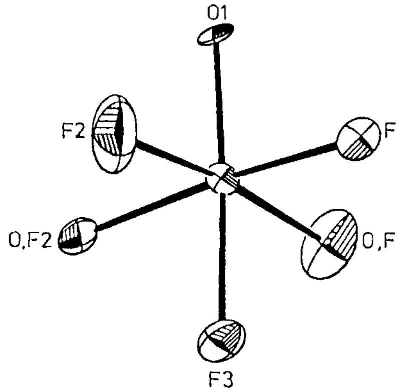

Figure 5. OsO_2F_4 molecular unit, ORTEP plot, ellipsoids show 50% probability. O,F refers to the disordered oxygen/fluorine atom pair. The atom O,F1 appears elongated along its bond direction, which is another indication of an O,F disorder. Selected bond lengths [pm]: Os—O(1) 164.2(25), Os—O,F(1,2) 180.5(23), 182.4(23), Os—F(1,2,3) 194.2(16), 195.2(36), 186.7(22); selected bond angles [°] O(1)—Os—O,F(1,2) 96.1(14), 94.4(11) F—Os—F(1 – 2, 1 – 3, 2 – 3) 83.8(9), 86.0(9), 83.0(15)

The previously mentioned *trans* effect should be observable here. But the accuracy of this crystal structure determination does not allow this, largely due to the fact that all atoms are superimpositions of two atoms, which also leads to abnormally large "vibrational" aplitudes of some of the ligand atoms. It cannot be excluded that the molecules are to a smaller extent oriented in even more than two ways, due to their spherical shape and lack of strong intermolecular interactions.

There are only a few compounds ($ReO_2F_4^-$, $WO_2F_4^{2-}$, $MoO_2F_4^{2-}$) which can be compared with OsO_2F_4, and all these have a *cis* orientation of oxygen atoms[3–6].

XeO_2F_4 is the only molecular analog of OsO_2F_4. But whether its structure is *cis* or *trans*, not to speak of other sophisticated details, is completely unknown. XeO_2F_4 has only been detected by its mass spectrum so far[47]. Among main group derivatives it is known that $IO_2F_4^-$ and $TeO_2F_4^{2-}$ exist both in the *cis* and *trans* configuration, and at present it cannot be decided with certainty which one is the more stable configuration[48,49]. A *trans* effect, that is a weakening of the fluorine bond *trans* to oxygen, has been discussed for *cis*-$TeO_2F_4^{2-}$, but precise structural details are not known[21].

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Experimental

General Procedure and Characterization: Volatile materials were manipulated in a metal vacuum line equipped with Teflon-FEP U tubes connected to the line by metal valves. Prior to handling the moisture-sensitive compounds, the system was passivated with chlorine trifluoride. The syntheses were performed in an 18 cm long, 13-mm wide sapphire tube or in a 6 mm wide Teflon-FEP tube equipped with metal valves. Moisture-sensitive non-volatile materials were transferred in the dry atmosphere of a glove box (Braun).

X-ray diffraction powder patterns: samples sealed in 0.5 mm wide quartz capillaries, Philips camera (diameter 11.46 cm) with Ni-filtered Cu-K_α radiation. – ^{19}F-NMR: Bruker Model MSL 300 instrument at 282.4 MHz, $CFCl_3$ external standard. – IR: Perkin Elmer Model 283. – Raman spectra: Coderg Model T 800 spectrometer, 647.1-nm excitation of a Kr ion Spectra Physics laser.

Materials: Osmium tetroxide OsO_4 from Merck was transferred and stored over P_2O_5. Chlorine trifluoride ClF_3 from Comurhex was purified by trap-to-trap distillation. Commercial HF (from Union Carbide) was dehydrated over bismuth pentafluoride before use[50]. Krypton difluoride KrF_2 was prepared as previously described[51].

Osmium Trioxide Difluoride, OsO_3F_2, was prepared according to a new method[52] by fluorination of OsO_4 with ClF_3. In a typical preparation, 1.1335 g (4.459 mmol) of OsO_4 was condensed at −196°C into the Teflon-FEP reaction tube. Then 0.784 g (8.482 mmol) of ClF_3 was added by condensation at the same temp. The solution, which turned red-orange upon warming to ambient temp., was kept at this temp. for 1 d. Besides OsO_3F_2, $OsOF_5$ was identified by Raman spectroscopy[53] among the reaction products. This compound which is more volatile than OsO_3F_2 was isolated by vaporization at −10°C and condensation into a trap cooled at −196°C. The yellow solid residue obtained after this separation was identified by its Raman spectrum[24], its X-ray diffraction powder pattern[13], and an elemental analysis as OsO_3F_2. The weights of OsO_3F_2 (1.0185 g) and $OsOF_5$ (0.210 g) amounted to 3.688 and 0.697 mmol, respectively. Thus, the yields of the conversion of OsO_4 into OsO_3F_2 and $OsOF_5$ were 82.7 and 15.6%, respectively. Single crystals of OsO_3F_2 were grown by sublimation in a static vacuum in a passivated sapphire tube. The temp. of the bottom of the sapphire tube was kept at 130°C.

Osmium Dioxide Tetrafluoride, OsO_2F_4, was obtained by fluorination of OsO_4 with KrF_2 in HF solution. The experimental conditions were similar to those used for the preparation of the new Os(VIII) oxide fluoride previously believed to be $OsOF_6$[8]. In a typical preparation 0.6023 g (2.369 mmol) of OsO_4 was allowed to react with 0.7536 g (6.187 mmol) of KrF_2 in 0.7102 g (35.51 mmol) of HF. Kyrpton difluoride was added by successive condensations till an excess of it was observed by Raman spectroscopy. The amount of the volatile components at −196°C was 0.5326 g, corresponding to 5.337 mmol of reacted KrF_2 if counted as a (1/2 O_2 + Kr) mixture versus 4.738 mmol expected for the conversion of the OsO_4 into OsO_2F_4. However, some decomposition of the KrF_2 cannot be ruled out so that the gaseous mixture could also contain some F_2. The volatile components at −78°C were evacuated in a dynamic vacuum at this temp. The solid residue was then left in the reaction tube inside the dry-box for 1 weeks to ensure decomposition of excess KrF_2. The solid was then pumped at −78°C for 4 h to eliminate F_2 and Kr. The obtained 0.7423 g of the red-brown solid corresponded to 2.489 mmol of OsO_2F_4, so some HF was still present. Part of this sample was used to record the ^{19}F-NMR spectrum, and the other part was purified by sublimation at 60°C in a static vacuum. The compound was extremely moisture-sensitive. – ^{19}F-NMR (HF solution, 25°C): $\delta = 11.5$, 59.6, $J_{FF} = 138$ Hz. – IR (solid): $\tilde{\nu} = 570$ cm^{-1} (sh), 588 (s), 675 (s), 930 (vs), 940 (vs). – Raman (solid): $\tilde{\nu} = 345$ cm^{-1} (30), 351 (33), 402 (33), 571 (15), 579 (18), 672 (60), 932 (32), 942 (100).

F_4O_2Os (298.2) Calcd. F 25.49 Os 63.78
Found F 25.63 Os 62.99

From HF solution:

$F_4O_2Os \cdot 2\,HF$ (338.2) Calcd. F 33.70 Os 56.24
Found F 33.53 Os 56.92

Single crystals of OsO_2F_4 were grown by sublimation in a static vacuum in a passivated sapphire tube, the bottom of which was kept at 40–50°C. The crystals were detached from the wall of the tube and selected in the dry box.

Table 1. Data collection, crystal data, and refinement results

	OsO_3F_2	OsO_2F_4
a [pm]	535.4(1)	489.3(2)
b [pm]	492.2(1)	489.3(2)
c [pm]	1208.7(2)	2719.0(1)
α [°]	90	90
β [°]	99.06(2)	90
γ [°]	90	120
V [10^6 pm^3]	315.8(2)	564.0(5)
T [°C]	−145	−154
Space group	$P2_1/c$	$P6_1$ (or $P6_5$)
Z	4	6
Crystal size [mm]	0.3 × 0.1 × 0.1	0.2 × 0.2 × 0.05
Color	yellow-brown	brown
μ [cm^{-1}]	425.9	358.9
Θ range measured [°]	2–40	2–40
Octands measured	$+h, +k, \pm l$	$+h, \pm k, +l$
Total no. of reflexions measured	1587	3624
No. of independent reflexions	1356	1279
No of reflexions used with $I \geq 3\sigma(I)$	1298	1169
R_{int}	0.033	0.033
No. of variable parameters	55	63
R	0.058	0.083
R_w	0.055	0.083
Weight scheme	6.5/σ2(*F*)	2.0/σ2(*F*)

Table 2. Atomic positional parameters and equivalent isotropic vibrational amplitudes for OsO_3F_2 and OsO_2F_4

Atom	x	y	z	B
Os	0.1934(1)	0.7143(1)	0.8641(0)	0.81
F(1)	-0.1308(16)	0.6035(17)	0.8865(7)	0.93
F(2)	-0.0406(17)	1.0339(17)	0.7897(7)	1.00
O(1)	0.4026(20)	0.8469(23)	0.7832(8)	1.23
O(2)	0.2484(22)	0.8948(24)	0.9840(8)	1.39
O(3)	0.3315(21)	0.4174(22)	0.9036(9)	1.20
Os	0.0023(3)	0.4589(2)	0.8333(0)	1.09
O(1)	0.2626(77)	0.7319(52)	0.7967(9)	1.89
F,O(1)	0.2858(50)	0.4494(87)	0.8748(11)	3.37
F,O(2)	-0.0548(54)	0.1260(51)	0.7956(7)	1.64
F(1)	-0.0472(50)	0.7581(46)	0.8739(7)	1.61
F(2)	-0.3530(92)	0.4393(89)	0.7975(15)	3.93
F(3)	-0.3177(58)	0.1462(58)	0.8718(8)	1.91

Crystal Structure Determinations: Suitable crystals were embedded into FEP oil, the suspension cooled to −100°C and mounted with the help of a special device[54,55] on an Enraf Nonius four-circle CAD 4 diffractometer. Lattice constants were refined by centering 25 standard reflexions, each having a Θ value between 12 and 25°. In the case of OsO_3F_2 the reflex intensities were measured with an ω-2Θ scan with a maximal measuring time of 80 s. After the Lorentz and polarization correction the structure was solved with the program SHELX 86[56]. Light atoms were found in difference Fourier maps, and finally all atoms were refined anisotropically. The absorption correction was performed according to the method of Walker and Stuart[57]. In the final refinement cycle the maximal shift versus the estimated standard deviation was 0.000. Two quite large residual electron densities were left at a distance of 90 pm from the osmium atom. For further results of the crystal structure determination see Tables 1 and 2.

Table 3. X-ray powder diffraction data of OsO_2F_4. s = strong, m = medium, w = weak, vw = very weak

d [Å]		Intensity	*hkl*	*d* [Å]		Intensity	*hkl*
Obsd.	Calcd.			Obsd.	Calcd.		
4.58	4.52	s	006	2.01	2.02	m	204
4.29	4.24	vw	100	1.84	1.97	m	205
	4.19		101				
4.10	4.05	s	102	1.94	1.86	mw	207
3.62	3.59	s	104	1.78	1.80	mw	208
3.12	3.09	w	106	1.68	1.67	vw	2010
2.68	2.65	m	108	1.62	1.61	w	2011
2.44	2.46	mw	109	1.59	1.60	w	211, 121
	2.45		110	1.58	1.58	w	213, 123
	2.44		111	1.57	1.57	mw	1016
	2.41		112	1.54	1.54	w	215, 125
2.33	2.30	mw	114	1.47	1.48	mw	217, 127
2.31	2.28	m	1010				
2.15	2.15	mw	116				
	2.13		1011				
2.10	2.09	vw	202				

OsO_2F_4 crystals appeared as very thin, hexagonal platelets. Thicker crystals were inevitably twinned. Even on the selected single crystal the reflex profile was 1.3° in ω and 0.4° in Θ. In connection with the longitudinal *c* axis the best compromise of scanning was ω-Θ. 71 out of 3695 reflexions were omitted because of overlap in the background measurement. The hexagonal lattice in connection with a clear extinction $00l$, $l = 6n$, allowed space groups $P6_1$ (no. 169), $P6_5$ (no. 170), $P6_122$ (no. 178), or $P6_522$ (no. 179). The identity of the crystal with bulk material was confirmed by indexing of the powder diffraction data (Table 3). The position of the osmium atom was found by means of the program SHELXS 86[52]. Light atoms were found in difference Fourier maps. The atomic coordinates of the oxygen and fluorine atoms excluded space group $P6_122$ or $P6_522$, although the osmium atom is close to a special position of the latter space groups. After the absorption correction[53] all atoms were refined anisotropically. The largest shift versus the estimated standard deviation was again 0.000, the residual electron density was found in the vicinity of the osmium atom.

In the calculations the disordered atoms O,F were treated as oxygen atoms, fluorine atoms, or as each of one type. *R* values did not differ characteristically. In the final calculation O,F1 was treated as oxygen, O,F2 as fluorine atom, giving the best U_{eq} values.

Further details of the crystal structures may be obtained from Fachinformationszentrum Karlsruhe, Gesellschaft für wissenschaftlich-technische Information mbH, D-7514 Eggenstein-Leopoldshafen 2, upon quoting the number CSD-57045, the names of authors and the journal citation.

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