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NMR investigation of the lability and Lewis acidity of some organotellurium(IV) dihalides: crystal structure of (1,3-dihydro-2λ^4-benzotellurole-2,2-diyl) difluoride, $C_8H_8TeF_2$

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Abstract

Tellurium-125 NMR data indicate that organotellurium(IV) dihalides $C_8H_8TeX_2$ (X = F, Cl, Br, I) do not form adducts with halide ion to any appreciable extent in solution. All the halides are labile in solution and mixed-halide species C_8H_8TeXX' have been observed for all halide combinations except X = F, X′ = I. Crystals of $C_8H_8TeF_2$ are monoclinic, space group $P2/n$ with $Z = 4$, a 9.3667(12), b 9.3009(9), c 9.5581(13) Å and β 103.422(11)°. Each Te is bonded to the two carbons of the C_8H_8 group as well as to four fluorine atoms; two of these fluorine atoms bridge asymmetrically between adjacent tellurium atoms to give an overall two dimensional polymer.

Introduction

As part of a program of investigation of the chemistry of hypervalent tellurium compounds in solution and in the solid state we investigated some simple organotellurium(IV) dihalide compounds of the type $C_8H_8TeX_2$ (X = F, Cl, Br, I; C_8H_8 = *o*-xylene-α, α'-diyl). The first complex in the series, $C_8H_8TeI_2$, has low solubility in common organic solvents, and conductance measurements show it to be monomeric in acetone, dichloromethane, and methyl ethyl ketone, but ionic in dimethylformamide [1]. Recrystallization from 2-methoxyethanol gives two crystalline forms of the compound, both of which have distorted octahedral geometries [2,3]. The remaining members of the series, $C_8H_8TeX_2$ (X = F, Cl, Br), are prepared by reaction of $C_8H_8TeI_2$ with AgX (X = F, Cl, Br). These compounds are stable crystalline solids which range in colour from orange (X = I) to colourless (X = F).

The tellurium-125 chemical shifts for $C_8H_8TeX_2$ (X = I, Cl, Br) and $C_8H_8TeR(X)$ (X = I, Br; R = alkyl) have been reported previously [4,5]. Halogen exchange in mixed dihalides also has been monitored previously by tellurium-125 NMR [4], but not all species were observed since the experiments were conducted at 32° C where apparently the rate of halide exchange is close to the NMR timescale, and this resulted in extremely broad and poorly resolved resonances which were difficult to assign. We now report results of our ^{125}Te NMR spectroscopic study of $C_8H_8TeX_2$ and the mixed halide compounds C_8H_8TeXX' in solution, as well the crystal structure of $C_8H_8TeF_2$.

Experimental

NMR spectra were recorded using a JEOL FX 100 spectrometer, generally with broad band proton decoupling. A Jeol NM 5471 controller was used for temperature control, the temperatures in the probe were measured with a platinum resistance thermometer. ^{125}Te spectra were recorded at 31.4 MHz usually on a 20 kHz spectral window; pulse width was 22 μs and pulse delay 50 ms. Spectra were recorded in the presence of $Cr(acac)_3$ to reduce relaxation times. ^{125}Te chemical shifts are relative to external 0.7 *M* K_2TeO_3 in water.

Preparation of the complexes $C_8H_8TeX_2$ (X = I, Br, Cl, F)

The complex $C_8H_8TeI_2$ was prepared as described by Ziolo and Günther [1]. The complexes $C_8H_8TeX_2$ (X = F, Cl, Br) were prepared by the reaction shown in general equation (eq. 1).

$$C_8H_8TeI_2 + 2AgX \rightarrow C_8H_8TeX_2 + 2AgI \qquad (1)$$

In a typical experiment 1.5 g $C_8H_8TeI_2$ (3 mmol) was stirred together with 1.0 g AgCl (7 mmol) in 200 cm^3 tetrahydrofuran for several hours or until the orange colour of $C_8H_8TeI_2$ was discharged. The solution was filtered and then allowed to evaporate slowly. Large colourless crystals of $C_8H_8TeCl_2$ were obtained in almost quantitative yield.

Crystallography

The crystal was mounted on a CAD-4F single crystal, four-circle, automatic diffractometer. Accurate cell dimensions were obtained from the setting angles of 25 reflections by a least-squares procedure. A pre-scan of $C_8H_8TeF_2$ showed the crystals to be monoclinic, with systematic absences consistent with the space groups $P2/n$ or Pn, which are the non-standard settings of the space groups $P2/c$ or Pc [6]. A satisfactory solution was achieved in the centrosymmetric space group, the non-standard setting of $P2/n$ being retained for the structure analysis. Intensity data were collected using the $\omega : 2\theta$ scan method to a maximum Bragg angle of 28.0°, using Mo-K_α radiation. Three reflections, which were monitored every 4000 s X-ray exposure time, indicated an 8% decrease in intensity during the data collection. The data were corrected for these intensity changes and for Lorentz and polarization effects. A total of 7784 reflections were measured, of which 1953 were unique, R_{amal} 0.034, and 1709 were considered observed, $I \geq 2\sigma(I)$.

The positions of the two crystallographically independent tellurium atoms were found from a three-dimensional Patterson map. The other non-hydrogen atoms

Table 1

Final fractional atomic coordinates for $C_8H_8TeF_2$

Atom	x	y	z
Te(1)	0.25000(–)	0.02300(3)	0.25000(–)
F(1)	0.2927(3)	0.0023(3)	0.0535(3)
C(1, 1)	0.0943(4)	−0.1408(3)	0.1837(5)
C(1, 2)	0.1751(3)	−0.2827(3)	0.2169(3)
C(1, 3)	0.1014(4)	−0.4124(3)	0.1842(4)
C(1, 4)	0.1758(4)	−0.5415(4)	0.2168(4)
Te(2)	−0.25000(–)	0.13368(3)	0.25000(–)
F(2)	−0.0528(2)	0.1560(2)	0.2020(3)
C(2, 1)	−0.1809(4)	0.2975(3)	0.4022(4)
C(2, 2)	−0.2167(3)	0.4386(3)	0.3227(3)
C(2, 3)	−0.1848(4)	0.5683(4)	0.3960(4)
C(2, 4)	−0.2177(4)	0.6973(4)	0.3223(4)

were located from subsequent difference maps. The structure was refined using a full-matrix least-squares refinement procedure, with anisotropic temperature factors assigned to all atoms. All hydrogen atoms were located from the difference maps, and were constrained at geometrical estimates with a C–H bond length of 1.08 Å. Refinement was continued with weighting schemes of type $w = k(\sigma^2(F) + g\,F^2)^{-1}$, where k and g were varied during the refinement. It was then noted that there were several intense, low-order, reflections which showed the effects of extinction. The structure was refined using a refinable isotropic extinction parameter, x, such that $F^\star{}_{\text{calc}} = F_{\text{calc}}(1 - (0.0001 \times F^2)/\sin\theta))$. The refinement of $C_8H_8TeF_2$ converged with R 0.027, R_w 0.028, k 4.874, g 0.0020, and x 0.0021(2). Final fractional atomic coordinates are given in Table 1.

Calculations were carried out using the programs SHELX-76 [7], ORTEP [8], DISTAN [9] and MEAN PLANES [10] on a VAX 11/780 computer. Scattering curves for atomic H, C, N, and S were those collected by Sheldrick [7], while those of Te and I were taken from ref. 11, the values being corrected for the real and imaginary dispersion terms [12]. Tables of least-squares planes, anisotropic thermal parameters for heavy atoms, H atom coordinates, and lists of calculated and observed structure factors may be obtained from the authors.

Crystal data: $C_8H_8F_2Te$, M 269.74, monoclinic, $P2/n$ [no. 13. C_{2h}^4], a 9.3667(12), b 9.3009(9), c 9.5581(13) Å, β 103.422(11)°, U 809.95Å^3, $Z = 4$, D_c 2.21 g cm^{-3}, D_m 2.21 g cm^{-3}, Mo-K_α (graphite monochromatized) radiation λ 0.71069 Å, crystal dimensions, ±(001) 0.067, ±(1$\bar{1}$1) 0.100, ±(110) 0.150 mm., absorption coefficient 36.41 cm^{-1}, max. and min. transmission factors, 0.6433 and 0.3538, F(000) 504.

Results and discussion

Solution NMR studies of organotellurium(IV) halides

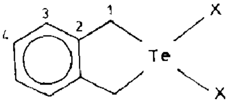
The class of compounds $C_8H_8TeX_2$ (X = F, Cl, Br, I), with the exception of $C_8H_8TeF_2$, is virtually insoluble in most non-coordinating organic solvents. They are, however, soluble in dimethylformamide (dmf), and NMR experiments were carried out at −40 °C to slow halide exchange relative to the NMR timescale so that sharp resonances could be observed. The NMR parameters of the dihalides and

Table 2

NMR data for $C_8H_8TeX_2$ (X = F, Cl, Br, I) in dmf solution at −40 °C.

Compound	$\delta(^{125}Te)$ [a]	$\delta(^{19}F)$	$\delta(^{13}C)$			
			1	2	3	4
$C_8H_8TeI_2$	−901	–	48.0	140.2	127.7	130.1
$C_8H_8TeBr_2$	−774	–	51.3	139.8	127.7	130.5
$C_8H_8TeCl_2$	−718	–	53.5	139.7	127.8	130.7
$C_8H_8TeF_2$	−378	−134.0	52.8	141.0	128.2	131.4
	J(Te–F) 1030	J(F–Te) 1030	^2J(C–F) 10 Hz			
C_8H_8TeIBr	−828	–				
C_8H_8TeICl	−784	–				
$C_8H_8TeBrCl$	−740	–				
C_8H_8TeFCl	−471	−137				
	J(Te–F) 814 Hz	J(F–Te) 815 Hz				
C_8H_8TeFBr	−453	−139				
	J(Te–F) 708 Hz	J(F–Te) 708				

[a] Relative to 0.7 M K_2TeO_3 in H_2O.



mixed halides are listed in Table 2. The tellurium-125 chemical shifts move progressively to higher frequency as the halogen becomes increasingly electronegative. The tellurium-125 resonance for $C_8H_8TeF_2$ is a triplet (J(Te–F) 1030 Hz), indicating that both fluorine atoms are equivalent and are bonded to the tellurium centre. Two-bond fluorine coupling is observed in the methylene carbon-13 triplet resonance (^2J(C–F) 10 Hz) in $C_8H_8TeF_2$. The methylene carbon-13 resonance appears to shift to higher frequency as the electronegativity of the halogen increases. The other aromatic carbon-13 resonances are little influenced by the nature of the halogen.

In most cases mixed halide compounds are observed in solution upon mixing the two corresponding dihalides. A tellurium-125 spectrum of a dmf solution containing equimolar proportions of $C_8H_8TeX_2$ and $C_8H_8TeX'_2$ show three resonances of relative intensities 1 : 2 : 1. The outer two resonances are at chemical shift positions for $C_8H_8TeX_2$ and $C_8H_8TeX'_2$ whilst the most intense resonance is assigned to the mixed ligand species C_8H_8TeXX' (eq. 2).

$$2C_8H_8TeX_2 + 2C_8H_8TeX'_2 \rightarrow C_8H_8TeX_2 + 2C_8H_8TeXX' + C_8H_8TeX'_2 \qquad (2)$$

The minimum requirements for this redistribution to occur are that at least one halide on each tellurium centre must be labile and that this lability is slow on the NMR timescale. The species C_8H_8TeFBr and C_8H_8TeFCl both give doublet tellurium-125 resonances indicating that the fluorine atom exchange is slow on the NMR timescale. As with the dihalides, the tellurium-125 chemical shifts of the mixed halide species move to higher frequency as the combined electronegativity of the halides increases.

Not all mixed halide combinations are observed in statistical distribution. Mixing equimolar proportions of $C_8H_8TeI_2$ and $C_8H_8TeCl_2$ in dmf results in formation of only a small amount of the mixed halide species, presumably owing to the limited solubility of C_8H_8TeICl in dmf. When equimolar proportions of $C_8H_8TeF_2$ and $C_8H_8TeI_2$ are mixed, only the two tellurium-125 resonances corresponding to the starting materials are observed, and there is no evidence for formation of C_8H_8TeIF.

It had earlier been reported that $C_8H_8TeI_2$ forms adducts of the type $[R_4N][C_8H_8TeI_2X]$ (where R = Me, Et, Bu; X = Cl, Br) in the solid state, but these adducts were only characterised by elemental analysis [13]. We also isolated the compound of stoichiometry $[Bu_4N][C_8H_8TeI_2Br]$ in order to characterise the species in solution by carbon-13 and tellurium-125 NMR. No tellurium-125 resonance could be found for a sample of the compound in dichloromethane (dcm) solution, probably because of the limited solubility in this solvent. The same compound in dmf solution gives three tellurium-125 resonances which correspond to the species $C_8H_8TeI_2$, C_8H_8TeIBr and $C_8H_8TeBr_2$. No other tellurium-125 resonances are observed, and it appears that the adduct $[C_8H_8TeI_2Br][Bu_4N]$ disproportionates in solution, probably by dissociation of a halide ion. We also isolated and studied the complex $[Bu_4N][C_8H_8TeI_3]$ [13]. The tellurium-125 spectrum of a solution of $[Bu_4N][C_8H_8TeI_3]$ in dmf at −40 °C, consists of a single resonance at $\delta(^{125}Te)$ −901 ppm which is attributed to $C_8H_8TeI_2$.

In order to confirm these results, the reactions of $C_8H_8TeI_2$ with Bu_4NX (X = I, Br) were performed in situ. A solution of $C_8H_8TeI_2$ in dmf gives a single tellurium-125 resonance at $\delta(^{125}Te)$ −901 ppm. Addition of an equimolar equivalent of Bu_4NI causes the tellurium-125 resonance to shift only slightly (i.e. to $\delta(^{125}Te)$ −905 ppm). Addition of a second equimolar equivalent of Bu_4NI causes a further small shift in the position of the tellurium-125 resonance (to $\delta(^{125}Te)$ −908 ppm). This minor chemical shift variation of the tellurium-125 resonances implies that the adduct $[C_8H_8TeI_3]^-$ is not formed to any appreciable extent in dmf.

The tellurium-125 spectrum of an equimolar mixture of $C_8H_8TeI_2$ and Bu_4NBr in dmf solution consists of three resonances at chemical shift positions identical to those observed in the equimolar mixture of $C_8H_8TeI_2$ and $C_8H_8TeBr_2$ in dmf at −40 °C, and indicates that there is no appreciable adduct formation under these conditions. The tellurium-125 spectrum of an equimolar solution of $C_8H_8TeF_2$ with Bu_4NF in dcm at −40 °C consists of a triplet, centred at $\delta(^{125}Te)$ −396 ppm, and two other very broad resonances at $\delta(^{125}Te)$ −480 and −539 ppm. It could not be determined whether the latter two resonances were due to a single tellurium species coupling to one fluorine atom or two unrelated tellurium containing compounds. The fluorine-19 spectrum contained several signals, none of which had tellurium-125 satellites. Both the tellurium-125 and fluorine-19 spectra are relatively unchanged in the temperature range 30 °C to −110 °C.

Description of the structure of $C_8H_8TeF_2$

An ORTEP diagram showing the atomic arrangement and the numbering scheme employed is given in Fig. 1. Relevant bond lengths and angles for $C_8H_8TeF_2$ are given in Table 3. The structure comprises two crystallographically unique tellurium atoms, each situated at a site of symmetry 2 and each of which is bonded to a *o*-xylene-α,α'-diyl group and four fluorine atoms. It can be seen from Table 3 that there are two distinct types of Te–F bond lengths. The shorter fall in the range 2.015(2) to 2.018(3) Å with the longer being 3.030(2) to 3.104(3) Å in length. These longer fluorine bonds bridge asymmetrically between adjacent tellurium atoms to

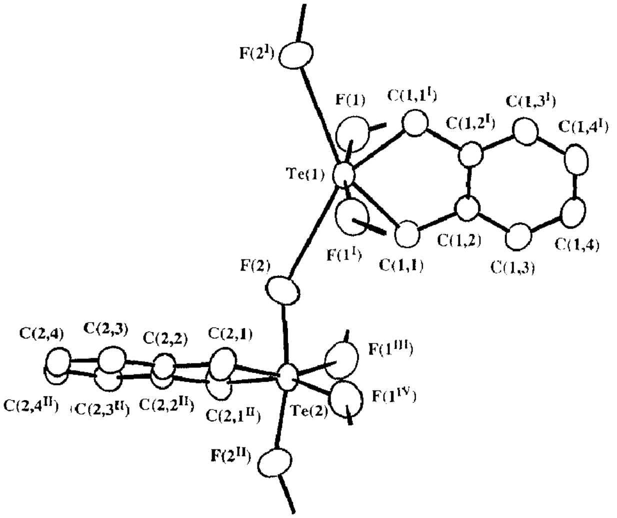


Fig. 1. ORTEP diagram of $C_8H_8TeF_2$ showing the numbering scheme employed.

Table 3

Important interatomic distances (Å) and angles (°) for $C_8H_8TeF_2$

Te(1)–C(1, 1)	2.103(3)	Te(2)–C(2, 1)	2.102(3)
Te(1)–F(1)	2.018(3)	Te(2)–F(2)	2.015(2)
Te(1)–F(2)	3.030(2)	Te(2)–F(1^{IV})	3.104(3)
C(1, 1)–C(1, 2)	1.518(4)	C(2, 1)–C(2, 2)	1.515(4)
C(1, 2)–C(1, 3)	1.389(4)	C(2, 2)–C(2, 3)	1.392(5)
C(1, 3)–C(1, 4)	1.387(5)	C(2, 3)–C(2, 4)	1.389(5)
C(1, 1)...C(1, 1^{I})	2.898(5)	C(2, 1)...C(2, 1^{II})	2.897(5)
C(1, 2)–C(1, 2^{I})	1.399(4)	C(2, 2)–C(2, 2^{II})	1.385(4)
C(1, 4)–C(1, 4^{I})	1.388(5)	C(2, 4)–C(2, 4^{II})	1.374(5)
F(1)...F(2^{I})	3.286(4)	F(1)...F(2^{III})	3.261(4)
F(1)–Te(1)–F(1^{I})	169.05(11)	F(2)–Te(2)–F(2^{II})	168.17(8)
F(1)–Te(1)–F(2)	106.19(9)	F(2)–Te(2)–F(1^{IV})	109.19(9)
F(1)–Te(1)–F(2^{I})	78.41(9)	F(2)–Te(2)–F(1^{III})	75.83(9)
F(1)–Te(1)–C(1, 1)	85.63(14)	F(2)–Te(2)–C(2, 1)	86.08(12)
F(1)–Te(1)–C(1, 1^{I})	86.44(15)	F(2)–Te(2)–C(2, 1^{II})	85.35(12)
C(1, 1)–Te(1)–F(2)	72.00(10)	C(2, 1)–Te(2)–F(1^{IV})	72.33(10)
C(1, 1)–Te(1)–F(2^{I})	154.28(10)	C(2, 1)–Te(2)–F(1^{III})	153.40(11)
C(1, 1)–Te(1)–C(1, 1^{I})	87.13(13)	C(2, 1)–Te(2)–C(2, 1^{II})	87.10(13)
F(2)–Te(1)–F(2^{I})	131.81(5)	F(1^{III})–Te(2)–F(1^{IV})	131.91(7)
Te(1)–C(1, 1)–C(1, 2)	106.8(2)	Te(2)–C(2, 1)–C(2, 2)	106.5(2)
C(1, 1)–C(1, 2)–C(1, 3)	120.7(3)	C(2, 1)–C(2, 2)–C(2, 3)	120.1(3)
C(1, 1)–C(1, 2)–C(1, 2^{I})	119.6(3)	C(2, 1)–C(2, 2)–C(2, 2^{II})	119.9(3)
C(1, 2)–C(1, 3)–(1, 4)	120.2(3)	C(2, 2)–C(2, 3)–C(2, 4)	119.8(3)
C(1, 3)–C(1, 2)–C(1, 2^{I})	119.7(3)	C(2, 3)–C(2, 2)–C(2, 2^{II})	119.9(3)
C(1, 3)–C(1, 4)–C(1, 4^{I})	120.0(3)	C(2, 3)–C(2, 4)–C(2, 4)	120.3(3)
Te(1)–F(1)–Te(2^{III})	153.91(13)	Te(1)–F(2)–Te(2)	143.03(10)

I $1/2-x,\ y, 1/2-z$ III $-x,\ -y,\ -z$

II $-1/2-x,\ y, 1/2-z$ IV $-1/2+x,\ -y, 1/2+z$

give an overall two dimensional polymer. The differences in dimensions between the two crystallographically unique molecules are minor and most likely due to packing forces.

Since the geometry about each of the unique tellurium atoms is almost identical, the following discussion is restricted to the geometry of the tellurium atom, Te(1). The coordination geometry formed by the four atoms closest to the tellurium atom, C(1, 1), C(1, 1^I), F(1), F(1^I), can be described as a distorted trigonal bipyramid, with the two fluorine atoms in the apical positions and two carbon atoms in the equatorial plane. The fifth coordination position, in the equatorial plane, is apparently occupied by a sterically active lone pair of electrons. The F(1)–Te(1)–F(1^I) angle of 169.05(11)° deviates considerably from linearity with both fluorine atoms pushed away from the equatorial lone pair. In addition, there are two further fluorine atoms, F(2) and F(2^I), which form secondary interactions with the tellurium atom and are smaller than the sum of the Van der Waals radii of 3.53 Å [14]. Inclusion of these two secondary interactions increases the coordination number of the tellurium atom to seven with one position occupied by a stereochemically active lone pair.

The structure of $C_8H_8TeF_2$ is very similar to that observed for α-$C_8H_8TeI_2$ [2]. The compound $C_8H_8TeI_2$ crystallizes in two crystalline forms, the structures of both α-$C_8H_8TeI_2$ and β-$C_8H_8TeI_2$ have been reported [2,3]. The carbon skeleton in all three structures is identical, although the tellurium–carbon bonds are shorter in $C_8H_8TeF_2$. Only slight differences occur in the angles subtended by the two shorter tellurium–halogen bonds. The I(1)–Te–I(2) angle in α-$C_8H_8TeI_2$ is 176.53(4)° with the same angle in β-$C_8H_8TeI_2$ being 179.53(3)°. In $C_8H_8TeF_2$ the corresponding angle, F(1)–Te(1)–F(1^I), is 169.05(11)°. This angle is smaller in $C_8H_8TeF_2$ than in α- or β-$C_8H_8TeI_2$ because of the shorter tellurium–fluorine bonds. A much larger difference is seen in the longer tellurium–halogen bonds. In $C_8H_8TeF_2$ the F(2)–Te(1)–F(2^I) angle is 131.81(5)°. The corresponding angle in α-$C_8H_8TeI_2$ is 111.96(3)°. This difference may also be explained due to the shorter tellurium–fluorine bonds, (i.e. Te(1)–F(2) 3.030(2) Å, Te(1)–F(2^I) 3.030(2) Å), as compared with the bonds in α-$C_8H_8TeI_2$, (i.e. Te–I(2^I) 3.653(1) Å, Te–I(2^{II}) 3.878(1) Å). In the compound β-$C_8H_8TeI_2$ however the angle I^{II}–Te–I^{III} is 66.87(2)°. Ziolo et al. describe both structures as having distorted octahedral geometries with no discussion of the stereochemical activity of the lone pair [2,3]. In β-$C_8H_8TeI_2$, the geometry is best described as a distorted octahedron with an inert lone pair. The small I^{II}–Te–I^{III} angle of 66.87(2)° is the result of an interaction between two iodine atoms, (i.e. $I^{II}\ldots I^{III}$ 4.042(1) Å). No iodine–iodine interactions are observed in α-$C_8H_8TeI_2$, but rather than being regarded as a distorted octahedron, with no reference to the large I(2^I)–Te–I(2^{II}) angle of 111.96(3)°, the structure can be better described in terms of a 1/2/2/2 geometry (including a stereochemically active lone electron pair).

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