

decreases slightly when the fraction of  $\text{CaCu}_5$ -type slabs increases. Little variation is observed in the  $C/a$  ratio. The situation is different for the members of the ternary series  $R_{2+n}T_{3+3n}M_{1+2n}$ . In fact, as mentioned above, the compression of the structure along the  $c$  axis is larger for the  $\text{CeCo}_3\text{B}_2$ -type slabs than for the ternary Laves-type slabs. As expected, the average thickness of the slabs is smaller the higher the fraction of  $\text{CeCo}_3\text{B}_2$ -type slabs in the structure. The  $a$  parameter seems to be determined essentially by the  $\text{CeCo}_3\text{B}_2$ -type slab and varies little when going from 50 to 100%  $\text{CeCo}_3\text{B}_2$ -type slabs. As a consequence,  $C/a$  decreases when  $n$  increases.

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## Hexatellurium Octaselenium Hexakis(hexafluoroarsenate) Sulfur Dioxide

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**Abstract.**  $(\text{Te}_6^{4+})(\text{Se}_8^{2+})(\text{AsF}_6^-)_6(\text{SO}_2)$ ,  $M_r = 2594.9$ , triclinic,  $P\bar{1}$ ,  $a = 12.407$  (4),  $b = 12.465$  (3),  $c = 14.109$  (5) Å,  $\alpha = 96.62$  (2),  $\beta = 90.49$  (3),  $\gamma = 95.13$  (2)°,  $U = 2158$  (1) Å<sup>3</sup>,  $Z = 2$ ,  $D_x = 3.99$  g cm<sup>-3</sup>, Mo  $K\alpha$  radiation ( $\lambda = 0.71069$  Å),  $\mu(\text{Mo } K\alpha) = 164.5$  cm<sup>-1</sup>,  $F(000) = 2276$ ,  $T = 298$  K,  $R = 0.063$  for 4896 reflections with  $F > 4\sigma(F)$ . The structure contains the previously known trigonal prismatic  $\text{Te}_6^{4+}$  cation

and the *exo-endo* cyclic  $\text{Se}_8^{2+}$  cation. The anion-cation interactions in the structure are discussed.

**Introduction.** In attempting to prepare new mixed Te–Se cations by the oxidation of mixtures of tellurium and selenium with  $\text{AsF}_5$  in  $\text{SO}_2$  as solvent, we have obtained a new dark green crystalline compound which has been shown by X-ray crystallography to contain

the two known cations  $\text{Te}_6^{4+}$  and  $\text{Se}_8^{2+}$  (Burns, Gillespie, Luk & Slim, 1979; McMullan, Prince & Corbett, 1971). This is the first time that two different homopolyatomic chalcogen cations have been found in the same crystalline compound.

**Experimental.** Anhydrous  $\text{SO}_2$  (30 mL) and  $\text{AsF}_5$  (2.37 g, 13.9 mmol) were distilled onto a mixture of powdered Te (0.4447 g, 3.485 mmol) and Se (0.2756 g, 3.490 mmol) in a double-bulb vessel. The initial green solution turned a deep blood-red color after about 0.5 h of stirring. This color persisted for 48 h, at which point the reaction vessel was heated to 338 K. After a few hours the solution became yellow-brown, and a large quantity of black crystalline material formed. Heating was continued for 1 week at 338 K with occasional stirring, but no further change was observed. On cooling to 298 K much of the black residue dissolved to give a deep green solution. In order to recrystallize the black material the reaction mixture was cooled to 195 K and then filtered. After 24 h at 298 K a large number of black crystals formed (ca 25% of the products). The composition of the remaining material was not determined. The solution was poured off and the crystals isolated under an atmosphere of  $\text{SO}_2$ . Under a microscope the rhombic-shaped crystals were observed to be dark green in color.

Crystals were selected and sealed in Lindemann capillaries in a dry box. Since the crystals tend to lose  $\text{SO}_2$  readily with a consequent loss of crystallinity, several crystals were studied before a suitable stable crystal was found from which a data set could be collected. Preliminary precession photographs were used to check crystal quality. Further work on a Syntex  $P2_1$  diffractometer using graphite-monochromatized  $\text{Mo K}\alpha$  radiation gave the experimental details summarized in Table 1. Final atomic positional parameters and bond angles in the  $\text{Te}_6^{4+}$  cation are given in Tables 2 and 3.\*

**Discussion.** Although we have reported several examples of mixed Se-Te cations, including  $\text{Te}_2\text{Se}_4^{2+}$ ,  $\text{Te}_2\text{Se}_6^{2+}$  and  $\text{Te}_2\text{Se}_8^{2+}$  (Boldrini, Brown, Gillespie, Ireland, Luk, Slim & Vekris, 1976; Boldrini, Brown, Collins, Gillespie, Maharajh, Slim & Sawyer, 1985; Gillespie, Luk, Maharajh & Slim, 1977; Collins, Gillespie & Sawyer, 1987) no mixed cations were obtained in the present study. Instead, the composition of the crystals was established as  $(\text{Te}_6)(\text{Se}_8)(\text{AsF}_6)_6(\text{SO}_2)$ . When the reaction was repeated at ca 298 K, precession photographs

Table 1. *Experimental details*

Number of reflections used in cell determination ( $2\theta$ range, °)	15 ( $27 < 2\theta < 30$ )
Mode	$\theta:2\theta$
Scan range (°)	( $K\alpha_1 - 0.9$ ) to ( $K\alpha_2 + 0.9$ )
Scan speeds (° min <sup>-1</sup> )†	3.5–29.3
Standard refin (number/interval)‡	3/67 refin
Max. $2\theta$ (°)/Quadrants	50/h, $\pm k, \pm l$
No. data collected §	8392 (incl. standards)
Structure solution	Direct methods ( <i>SHELX76</i> ; Sheldrick, 1976), Fourier, least squares
$h, k, l$ range	0/14, $-14/14, -16/16$
Crystal faces: $d$ (mm)	{(111), (111)} 0.075 {(111), (111)} 0.15 {001} 0.11
Absorption correction¶	
Grid/ $A^*$ values	$8 \times 8 \times 9/6.155-24.017$
No. data $F > 4\sigma(F)$	4896
$R(wR)\dagger\dagger$	0.063 (0.068)
Max. shift/e.s.d.	0.10
Weights	$[\sigma^2(F) + 0.00085F^2]^{-1}$
Final $\Delta F$ Fourier max. peak ( $e \text{ \AA}^{-3}$ )	$2.71(1.35 \text{ \AA} \text{ from Se}(4))$ $\pm 2.0$ (near Te atoms)
Scattering factors	<i>International Tables for X-ray Crystallography</i> (1974)
Programs	<i>XRAY76</i> (Stewart, Machin, Dickinson, Ammon, Heck & Flack, 1976) and <i>SHELX</i> on CDC6400 computer

† Dependent on intensity of a prescan. Stationary background counts measured at each end of scan, each for one quarter of scan time.

‡ No systematic variations in intensities observed.

§ Lorentz and polarization corrections applied to all data. Absorption corrections applied after composition of crystal had been established.

¶ Calculations gave  $R = 0.121$  for least-squares refinement using 4187 reflections with  $F > 6\sigma(F)$  before any absorption corrections and  $R = 0.058$  for the same refinement using the absorption-corrected data.

†† Block-diagonal (four blocks) least squares minimizing  $\sum w\Delta F^2$ . Structure factor calculation for 5857 data with  $F > 2\sigma(F)$  gave  $R = 0.080$ .

revealed that  $(\text{Te}_2\text{Se}_4)(\text{AsF}_6)_2$  was the only crystalline product, while the  $^{77}\text{Se}$  NMR spectrum of the mother liquor (Collins, 1984) identified the  $\text{Te}_2\text{Se}_4^{2+}$ ,  $\text{Te}_2\text{Se}_3^{2+}$  and *cis*- $\text{Te}_2\text{Se}_2^{2+}$  cations (Burns, Granger & Schrobilgen, 1978). The  $^{77}\text{Se}$  NMR spectrum of  $(\text{Te}_6)(\text{Se}_8)(\text{AsF}_6)_6(\text{SO}_2)$  redissolved in  $\text{SO}_2$  consisted of the five resonances due to the  $\text{Se}_8^{2+}$  cation (Burns, Collins, Gillespie & Schrobilgen, 1986). After several weeks at 298 K, however, the  $^{125}\text{Te}$  NMR spectrum of this sample revealed that much of the  $\text{Te}_6^{4+}$  had reacted to form  $\text{Te}_2\text{Se}_4^{2+}$ . Thus it would appear that an equilibrium is established in solution between  $\text{Te}_6^{4+}$  and  $\text{Se}_8^{2+}$  and a number of mixed cations. Perhaps the equilibrium shifts in favor of the homopolyatomic cations with increasing temperature and it appears that  $(\text{Te}_6)(\text{Se}_8)(\text{AsF}_6)_6(\text{SO}_2)$  has a lower solubility than the corresponding salts of the mixed cations at elevated temperatures.

The present  $\text{Te}_6^{4+}$  cation has a slightly distorted trigonal prismatic structure in which the end triangular faces are eclipsed with a small but significant angle of  $3.00^\circ$  between their planes (Fig. 1). This results in

\* Lists of structure-factor amplitudes, anisotropic thermal parameters and bond lengths/bond angles for the  $\text{AsF}_6^-$  anions and  $\text{SO}_2$  solvent, and Fig. 4 showing contacts to the  $\text{Se}_8^{2+}$  cation have been deposited with the British Library Document Supply Centre as Supplementary Publication No. SUP 44489 (37 pp.). Copies may be obtained through The Executive Secretary, International Union of Crystallography, 5 Abbey Square, Chester CH1 2HU, England.

Table 2. Final atomic positional parameters ( $\times 10^4$ ) and isotropic or equivalent isotropic thermal parameters ( $\text{\AA}^2 \times 10^3$ )

	$U_{\text{eq}} = \frac{1}{3} \text{trace } U$			
	x	y	z	U or $U_{\text{eq}}$
Te(1)	2432 (1)	4198 (1)	304 (1)	41.8 (5)
Te(2)	831 (1)	2603 (1)	282 (1)	43.3 (5)
Te(3)	1616 (1)	3189 (1)	-1381 (1)	43.9 (5)
Te(4)	3280 (1)	1530 (1)	-1422 (1)	42.0 (5)
Te(5)	2568 (1)	880 (1)	243 (1)	41.1 (5)
Te(6)	4165 (1)	2477 (1)	256 (1)	41.8 (5)
Se(1)	5869 (2)	2105 (2)	5369 (2)	48.0 (8)
Se(2)	6995 (2)	2770 (2)	6638 (2)	56.1 (9)
Se(3)	7968 (2)	1288 (2)	6527 (2)	70.8 (11)
Se(4)	8954 (2)	1950 (2)	5303 (2)	70.7 (11)
Se(5)	7625 (2)	1428 (2)	4158 (2)	51.5 (8)
Se(6)	7719 (3)	2887 (2)	3281 (2)	77.9 (12)
Se(7)	7455 (2)	4250 (2)	4492 (2)	64.1 (10)
Se(8)	5654 (2)	3649 (2)	4629 (2)	62.1 (10)
As(1)	4719 (2)	4645 (2)	-2035 (1)	33.7 (6)
As(2)	7505 (2)	2507 (2)	-82 (2)	37.6 (7)
As(3)	-167 (2)	196 (2)	2033 (1)	34.1 (6)
As(4)	4766 (2)	543 (2)	2462 (2)	38.7 (7)
As(5)	2333 (2)	1947 (2)	-4287 (2)	44.0 (7)
As(6)	288 (2)	5062 (2)	2528 (2)	42.1 (7)
S	2469 (6)	2974 (6)	3141 (4)	73 (3)
O(1)	2575 (12)	2818 (13)	2141 (10)	62 (6)
O(2)	3086 (15)	3847 (16)	3655 (12)	88 (8)
F(11)	6065 (10)	4647 (13)	-1855 (12)	90 (7)
F(12)	4601 (12)	3251 (10)	-2178 (11)	78 (6)
F(13)	4523 (12)	4583 (13)	-828 (9)	85 (7)
F(14)	3344 (10)	4612 (11)	-2161 (10)	72 (6)
F(15)	4870 (11)	4674 (11)	-3223 (9)	71 (5)
F(16)	4852 (12)	6011 (9)	-1832 (11)	83 (6)
F(21)	6581 (15)	3407 (15)	-9 (14)	121 (6)*
F(22)	8451 (16)	1607 (16)	-131 (15)	134 (7)*
F(231)†	8500 (27)	3468 (26)	-414 (26)	93 (10)*
F(241)†	6518 (26)	1528 (26)	249 (26)	89 (10)*
F(251)†	7810 (37)	2791 (35)	1098 (32)	135 (15)*
F(261)†	7246 (34)	2079 (35)	-1220 (30)	124 (12)*
F(232)†	7774 (39)	2988 (39)	-1109 (35)	147 (16)*
F(242)†	6931 (43)	2161 (41)	911 (38)	163 (18)*
F(252)†	6630 (30)	1569 (29)	-614 (30)	108 (11)*
F(262)†	8371 (31)	3470 (30)	443 (30)	114 (12)*
F(31)	-1498 (10)	145 (12)	2249 (9)	75 (6)
F(32)	134 (11)	440 (13)	3188 (9)	81 (6)
F(33)	-469 (11)	-117 (13)	829 (9)	76 (6)
F(34)	1169 (10)	223 (13)	1763 (10)	81 (6)
F(35)	-216 (13)	-1161 (10)	2082 (12)	86 (7)
F(36)	-135 (14)	1516 (11)	1877 (13)	102 (8)
F(41)	6164 (9)	626 (10)	2538 (10)	64 (5)
F(42)	4881 (11)	185 (12)	1236 (9)	73 (6)
F(43)	4729 (10)	-796 (10)	2584 (11)	69 (6)
F(44)	4712 (13)	892 (16)	3640 (10)	106 (8)
F(45)	3404 (9)	464 (11)	2342 (10)	71 (6)
F(46)	4825 (12)	1869 (10)	2250 (14)	96 (7)
F(51)	1507 (14)	2656 (14)	-4859 (13)	107 (6)*
F(52)	3105 (18)	1295 (18)	-3595 (17)	148 (8)*
F(53)	1345 (17)	954 (16)	-4241 (15)	134 (7)*
F(54)	3390 (15)	2818 (15)	-4432 (14)	121 (6)*
F(55)	2002 (22)	2562 (21)	-3258 (20)	188 (11)*
F(56)	2645 (23)	1310 (24)	-5273 (22)	206 (12)*
F(61)	-1075 (9)	4999 (11)	2630 (10)	68 (5)
F(62)	102 (12)	5090 (11)	1318 (10)	75 (6)
F(63)	1660 (10)	5129 (11)	2329 (11)	78 (6)
F(64)	336 (10)	6451 (9)	2668 (11)	67 (5)
F(65)	251 (11)	3698 (10)	2313 (12)	78 (6)
F(66)	484 (16)	5015 (15)	3693 (12)	111 (9)

\* Refined isotropically.

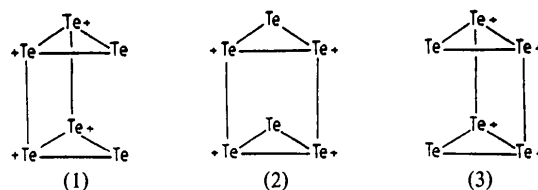
† Refined as half a fluorine atom.

Table 3. Bond angles ( $^\circ$ ) in the  $\text{Te}_6^{4+}$  cation

Te(2)-Te(1)-Te(3)	60.25 (5)	Te(3)-Te(4)-Te(5)	91.48 (6)
-Te(6)	90.17 (5)	-Te(6)	91.43 (5)
Te(3)-Te(1)-Te(6)	88.81 (5)	Te(5)-Te(4)-Te(6)	59.48 (5)
Te(1)-Te(2)-Te(3)	60.28 (5)	Te(2)-Te(5)-Te(4)	88.51 (6)
-Te(5)	89.82 (5)	-Te(6)	90.08 (5)
Te(3)-Te(2)-Te(5)	88.84 (6)	Te(4)-Te(5)-Te(6)	60.19 (5)
Te(1)-Te(3)-Te(2)	59.47 (5)	Te(1)-Te(6)-Te(4)	88.64 (6)
-Te(4)	91.13 (6)	-Te(5)	89.93 (5)
Te(2)-Te(3)-Te(4)	91.17 (6)	Te(4)-Te(6)-Te(5)	60.32 (5)

Te-Te-Te angles of 88.51 to 91.48 (6) $^\circ$  in the 'rectangular' faces. Although the cation in  $(\text{Te}_6)(\text{AsF}_6)_4(\text{SO}_3)_2$  has no crystallographic symmetry, it is the most regular example of this cation since the end triangular faces are parallel to within 0.60 $^\circ$ . In comparison, the cation in  $(\text{Te}_6)(\text{AsF}_6)_4(\text{AsF}_3)_2$  has crystallographic symmetry 2 and an angle of 1.75 $^\circ$  between the end triangular faces which are also twisted by 2.7 $^\circ$  with respect to each other (Burns *et al.*, 1979).

In  $(\text{Te}_6)(\text{AsF}_6)_4(\text{SO}_3)_2$  all the bonds in the end triangular faces have the same length as do the bonds between these faces. In the present cation the bonds between the triangular faces have lengths of 3.047 (2), 3.167 (2) and 3.172 (2)  $\text{\AA}$ . In each end triangular face the bonds which include the Te atoms involved in the short bond between these faces are significantly longer than the remaining bond. A less pronounced correlation is observed for the Te-Te bond lengths in  $(\text{Te}_6)(\text{AsF}_6)_4(\text{AsF}_3)_2$ . Since the bonds in the end triangular faces are shorter than those between these faces the bonding in the  $\text{Te}_6^{4+}$  ion may be approximately described in terms of the resonance structures (1)-(3).



The present  $\text{Se}_8^{2+}$  cation is only very slightly distorted from  $C_s$  symmetry and has the same *exo-endo* configuration as that observed in  $\text{Se}_8(\text{AlCl}_4)_2$  and the isostructural  $\text{S}_8^{2+}$  cations in  $\text{S}_8(\text{AsF}_6)_2$  and  $\text{S}_8(\text{Sb}_3\text{F}_{14})(\text{SbF}_6)$  (Davies, Gillespie, Park & Passmore, 1971; Gillespie, Sawyer & Vekris, unpublished results). Bond lengths and bond angles in the present cation are compared with those for  $\text{Se}_8(\text{AlCl}_4)_2$  in Fig. 2. Unlike the cation  $\text{Se}_8^{2+}$  (Burns, Chan, Gillespie, Luk, Sawyer & Slim, 1980) where there is a significant bond-length alternation on moving away from the positively charged atoms, the bonds in the  $\text{Se}_8^{2+}$  cation do not show any

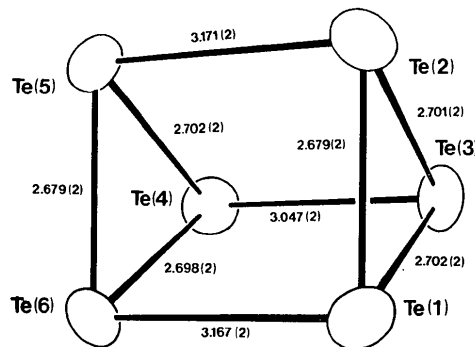


Fig. 1. ORTEP (Johnson, 1965) diagram showing the principal dimensions of the  $\text{Te}_6^{4+}$  cation in  $(\text{Te}_6)(\text{Se}_8)(\text{AsF}_6)_6(\text{SO}_3)_2$ .

alternation in length around the ring, although there are some significant differences between individual bond lengths. The  $\langle \text{Se}-\text{Se} \rangle$  bond length in  $\text{Se}_8^{2+}$  (2.306 Å) is shorter than the bond length of 2.34 Å in the crown-shaped  $\text{Se}_8$  ring (Foss & Janickis, 1980).

An important feature of the  $\text{Se}_8^{2+}$  structure is the short cross-ring  $\text{Se}(1)\cdots\text{Se}(5)$  distance of 2.905 (3) Å which is slightly longer than the analogous distance of 2.84 (1) Å in  $\text{Se}_8(\text{AlCl}_4)_2$  and is *ca* 0.9 Å less than twice the van der Waals radius for Se (3.8 Å). It is very similar in length to the long Se-Se bond in the  $\text{Se}_2\text{I}_4^{2+}$  cation (2.84 Å) (Nandana, Passmore, White & Wong, 1982). Other weakly bonding Se $\cdots$ Se interactions have been observed in  $\text{Se}_2^{2+}$  (3.30–3.49 Å),  $\text{Se}_4\text{S}_2\text{N}_4^{2+}$  (3.12–3.18 Å) and  $\text{Se}_4\text{N}_4$  (2.75 Å) (Gillespie, Kent & Sawyer, 1981; Barnighausen, Volkmann & Jander, 1966). In the present  $\text{Se}_8^{2+}$  cation the small angles of 90.0 (1)° for Se(2–3–4) and 92.7 (1)° for Se(6–7–8) are consistent with the cross-ring bonding.

One of the  $\text{AsF}_6^-$  anions in the asymmetric unit is disordered over two equal orientations related by a rotation of *ca* 45° about the F(21)–As(2)–F(22) axis. The large fluorine thermal parameters indicate that the anion  $\text{As}(5)\text{F}_6^-$  is only weakly held in the lattice. All six anions are fairly regular octahedra with As–F distances of 1.59–1.75 Å. Many of the longer bonds involve the F atoms forming the shortest Te $\cdots$ F and Se $\cdots$ F contacts. The  $\langle \text{S}-\text{O} \rangle$  distance of 1.40 (2) Å and

the OSO angle of 118 (1)° in the  $\text{SO}_2$  molecule are comparable with those found for crystalline  $\text{SO}_2$  and for  $\text{SO}_2$  molecules in other compounds (Burns *et al.*, 1979; Post, Schwartz & Fankuchen, 1952).

In  $(\text{Te}_6)(\text{Se}_8)(\text{AsF}_6)_6(\text{SO}_2)$  the crystal packing consists of layers of anions and cations parallel to the  $(\bar{1}01)$  plane (Fig. 3). Several interionic contacts to the  $\text{Te}_6^{4+}$  cation are comparable in length to the contacts in the other  $\text{Te}_6^{4+}$  salts and probably involve charge transfer from O or F atoms into antibonding orbitals of the cations. In  $\text{Te}_6^{4+}$  this charge transfer is probably into the LUMO which has symmetry  $A'_2$  assuming a regular  $D_{3h}$  geometry for the cation (Burns, Gillespie, Barnes & McGlinchey, 1982). The shorter contacts cap some of the rectangular and triangular faces or bridge edges of the cation and are formed towards presumed minima in the electron density around the cation. In the present structure, Te(3) and Te(4) which form the short bond in the rectangular face (and the long bonds in the triangular faces) form five or six Te $\cdots$ F contacts in the range 2.73–3.15 Å, while the other Te atoms have only two to three contacts of similar length.

The Se $\cdots$ F contacts to the  $\text{Se}_8^{2+}$  cation are fewer in number and probably weaker than the analogous Se $\cdots$ Cl contacts in  $\text{Se}_8(\text{AlCl}_4)_2$  (Fig. 4)\* which is consistent with the greater polarizability of Cl. Many of the Se $\cdots$ F and Se $\cdots$ Cl contacts are approximately collinear ( $\sim 160$ – $180^\circ$ ) with the Se–Se primary bonds or are approximately *trans* to the assumed directions of the lone pairs [*cf.* the arrangement of similar weak contacts to divalent S and Se atoms (Rosenfield, Parthasarathy & Dunitz, 1977; Gillespie *et al.*, 1981)]. The shortest Se $\cdots$ F and Se $\cdots$ Cl contacts are to the Se atoms linked by the transannular interaction which nominally carry the positive charge. If this interaction is considered to be a bond, there is an approximately mon capped octahedral  $AX_3Y_3E$  arrangement of bonds (X) and secondary Se $\cdots$ F (Se $\cdots$ Cl) interactions (Y) around these atoms.

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\* See deposition footnote.

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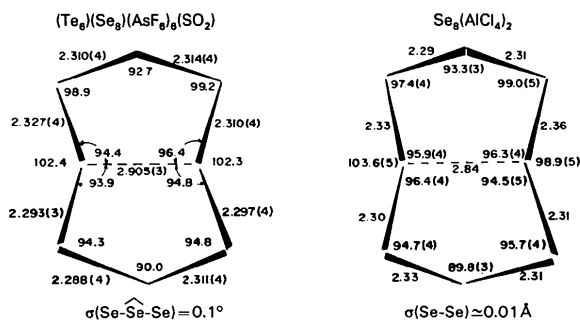


Fig. 2. Dimensions of the  $\text{Se}_8^{2+}$  cations in  $(\text{Te}_6)(\text{Se}_8)(\text{AsF}_6)_6(\text{SO}_2)$  and  $\text{Se}_8(\text{AlCl}_4)_2$ .

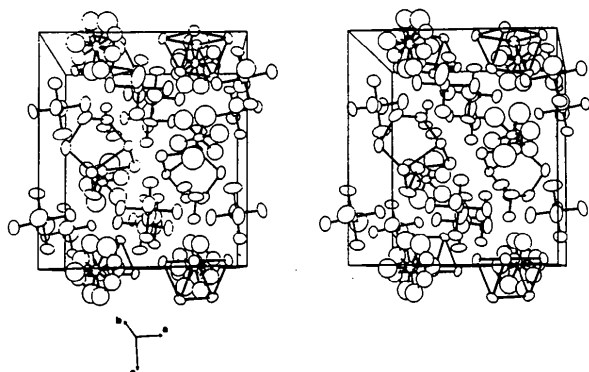


Fig. 3. Crystal packing as viewed down *b*.

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## The Structures of Tutton's Salts. I. Diammonium Hexaaquamagnesium(II) Sulfate

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**Abstract.** The crystal structure of the title compound at 295 K was refined using both neutron and X-ray data.  $[\text{NH}_4]_2[\text{Mg}(\text{H}_2\text{O})_6](\text{SO}_4)_2$ ,  $M_r = 359.28$ , monoclinic,  $P2_1/a$ ,  $a = 9.316$  (3),  $b = 12.596$  (6),  $c = 6.198$  (2) Å,  $\beta = 107.09$  (3)°,  $V = 695.22$  Å<sup>3</sup> (X-ray results),  $Z = 2$ ,  $D_x = 1.723$  Mg m<sup>-3</sup>,  $F(000) = 380$ . X-ray data: Mo  $K\alpha$ ,  $\lambda = 0.71069$  Å,  $\mu = 0.496$  mm<sup>-1</sup>,  $R = 0.065$  for 7212 reflections. Neutron data:  $\lambda = 0.9891$  Å,  $\mu = 0.21$  mm<sup>-1</sup>,  $R = 0.041$  for 1372 reflections. Covalent bond lengths in the structure are affected significantly by interactions between neighbouring groups. The differences between the neutron and X-ray structures also vary with the bonding environment, providing another measure of the strengths of these interactions.

**Introduction.** The title compound is a member of the isomorphous series  $(\text{NH}_4)_2M^{II}(\text{SO}_4)_2(\text{H}_2\text{O})_6$ , known as Tutton's salts.  $M$  is a divalent metal. The structural characteristics of the series, including details of the hydrogen-bond network, are described by Montgomery & Lingafelter (1966). The magnesium member, with  $M = \text{Mg}$ , is a convenient reference system, with which the complexes containing heavier metals may be compared. The X-ray scattering for magnesium is

strong enough to yield reliable thermal parameters, but not so large as to preclude a study of the effect of hydrogen bonding on the electron density near the H nuclei. The bias in the X-ray positions, revealed by comparison with a neutron diffraction structure, indicates a local dipole component in the redistribution of electron density due to bonding. Neutron and X-ray data were collected, and the structures refined with these data to study these local dipole terms, as the first stage of the study of the electron density. Previous structural work on the magnesium complex is described by Montgomery & Lingafelter (1964).

**Experimental.** Crystals by evaporation of a solution containing stoichiometric quantities of ammonium and magnesium sulfates in water. Neutron measurements at 295 K with neutrons of wavelength  $\lambda = 0.9891$  Å on the 4-circle diffractometer at the HIFAR reactor at the Australian Atomic Energy Commission Research Establishment at Lucas Heights, New South Wales. Crystal volume 46.0 mm<sup>3</sup>, 14 faces. Neutron cell dimensions determined by least-squares fit to the angular settings of 30 reflections fully centred in the counter aperture:  $a = 9.316$  (2),  $b = 12.580$  (4),  $c = 6.202$  (1) Å,  $\beta = 107.094$  (5)°.