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A Refinement of the Crystal Structure of Potassium Imidodisulphate

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The structure of potassium imidodisulphate, $K_2[NH(SO_3)_2]$, has been refined three-dimensionally in the space-group $C2/c$ by the method of least squares with allowance for anisotropic vibrations and corrections for rotational oscillations. Refinement to a disagreement index of 0·094 over 755 planes has led to the corrected bond lengths S–N = 1·662 ± 0·005 Å and mean S–O = 1·453 ± 0·005 Å, and bond angles S–N–S = 124·5°, mean O–S–O = 113·0° and mean N–S–O = 105·6°. The hydrogen atom was not explicitly located but is believed to be coplanar with the nitrogen and sulphur atoms. Comparisons are made with the related crystal structures of potassium pyrosulphate and potassium methylenedisulphate.

Introduction

The crystal structure of potassium imidodisulphate or iminodisulphonate (previously called aminedisulphonate), $K_2[NH(SO_3)_2]$, was determined by Jeffrey & Jones (1956) by three-dimensional Fourier methods, ending with an isotropic refinement by two cycles of observed and calculated differential syntheses. From these, the S–N bond was found to have a length 1·655 ± 0·007 Å; Jeffrey & Jones regarded this as a partial double bond as it was intermediate between the S–N lengths of 1·60 ± 0·03 Å in the amidosulphate ion, $[SO_3NH_2]^-$, and 1·79 ± 0·02 Å in the dinitrososulphite ion, $[SO_3N_2O_2]^{2-}$.

Since the structures of the isomorphous crystals of potassium methylenedisulphate, $K_2[CH_2(SO_3)_2]$ (Jones, 1955; Truter, 1962), and potassium pyrosulphate, $K_2S_2O_7$ (Lynton & Truter, 1960), have recently been determined by least-squares methods, with allowance for anisotropic vibrations and rotational-oscillation corrections, it seemed desirable to refine the imidodisulphate data by similar techniques so as to have available dimensions of comparable accuracy for the three isostructural salts. The refinement reported here leads to dimensions differing very little from those reported by Jeffrey & Jones; however, it is possible now to give a fuller description of the crystal structure and of the bonds.

Least-squares refinement

Potassium imidodisulphate crystallizes with the space group $C2/c$ in a unit cell of dimensions

$$a = 12{\cdot}430,\ b = 7{\cdot}458,\ c = 7{\cdot}175\ \text{Å};\ \beta = 91°\ 11'.$$

The eight K^+ cations are in general positions and the four $[NH(SO_3)_2]^{2-}$ anions are astride twofold axes passing through the nitrogen atoms and possibly through the hydrogen atoms, although the latter were not located by Jeffrey & Jones in an $(F_o - F_c)$ synthesis down the $(0, y, \frac{1}{4})$ axis.

The X-ray data used in this refinement were the 687 non-zero $|F_o|$ measured by Jeffrey & Jones together with 68 weak reflexions which were included with values equal to the locally observable minimum $|F_o|$. (As an indication of absorption and estimation errors, measurements from two sets of $\{hk0\}$ photographs taken from different crystals led to a discrepancy index in $|F_o|$ of 0·05 taken over all 64 reflexions recorded.) Five strong reflexions (020, 202, $\bar{2}02$, 400, and 021) were omitted from the least-squares minimization because of presumed extinction, and four others (listed at the end of Table 3) were excluded because of doubts as to their reliability.

Table 1. *Uncorrected monoclinic co-ordinates from least-squares refinements*

Atom	*x* (Å)	*y* (Å)	*z* (Å)
K	4·3201	4·7835	4·6115
S	4·9574	1·3296	4·5903
O(1)	5·5152	2·0264	3·4576
O(2)	4·0792	0·2500	4·2259
O(3)	4·3743	2·2364	5·5416
N	6·2150 (= *a*/2)	0·5745	5·3812 (= 3*c*/4)
H	6·2150 (= *a*/2)	−0·4300*	5·3812 (= 3*c*/4)

* Assumed.

All the calculations were carried out on the Leeds University Pegasus Computer with programs described by Cruickshank, Pilling, Bujosa, Lovell & Truter (1961). Instead of the Hartree scattering factors with separate isotropic thermal parameters used in the earlier differential syntheses, the scattering factors of Berghuis, Haanappel, Potters, Loopstra, MacGillavry & Veenendaal (1955) for K^+, O, and N, and a slightly improved version of the curve given by Tomiie & Stam (1958) for S were used in the least-squares refinement. After several cycles in which

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Table. 2. *Vibration tensor components with respect to crystal axes*
(All values in Å^2)

Atom	U_{11}	U_{22}	U_{33}	$2U_{12}$	$2U_{23}$	$2U_{13}$
K	0·0403	0·0417	0·0340	0·0008	0·0131	0·0026
S	0·0292	0·0323	0·0270	0·0022	0·0017	0·0043
O(1)	0·0420	0·0600	0·0386	−0·0125	0·0330	0·0039
O(2)	0·0390	0·0455	0·0547	−0·0088	−0·0081	−0·0076
O(3)	0·0440	0·0437	0·0489	0·0257	0·0027	0·0033
N	0·0374	0·0280	0·0332	0	0	0·0133

allowance was made for the anisotropic vibrations of K$^+$, S, and O, the *R* index fell from 0·120 to 0·104. An attempt was then made to locate the hydrogen atoms by carrying out parallel calculations from two sets of atomic positions differing only in the postulated hydrogen sites: (i) a single atom on the two-fold axis, corresponding to coplanarity of the N–H and two N–S bonds; and (ii) a statistical distribution of half hydrogen atoms. In neither case was the hydrogen position confirmed and so this atom was not refined in the full anisotropic refinement, of which nine cycles were carried out. These led to the final atomic coordinates shown in Table 1 and the anisotropic vibration parameters shown in Table 2; the largest co-ordinate change from the Jeffrey & Jones values was only 0·018 Å (in the *x* co-ordinate of oxygen O(1)). The final values of observed and calculated structure factors are listed in Table 3 and correspond to an *R* index of 0·094 in which unobserved planes are compared at their maximum possible values. The inclusion of the unobserved reflexions in the least-squares calculations in this way is not entirely satisfactory, but it is unlikely to have affected the results materially. In the later stages of the refinement, the weighting scheme used was

$$w = 1/(4\cdot0 + |F_o| + |F_o|^2/40)\ ,$$

where the $|F_o|$'s are on the scale used by Jeffrey & Jones. According to the least-squares refinement, this scale should be increased by a factor 1·064 ± 0·012.

The co-ordinates' e.s.d.'s determined by the least-squares process are virtually isotropic and have magnitudes 0·0016 Å for K, 0·0015 Å for S, 0·006 Å for O, and 0·008 Å for the *y* co-ordinate of N; the e.s.d.'s of the diagonal elements of the mean square vibration amplitudes, U_{ij}, are roughly 0·008 Å^2 for K and S, 0·0030 Å^2 for O, and 0·0036 Å^2 for N.

Molecular vibration analysis

The values of the U_{ij} in Table 2 confirm and elaborate the observation from the earlier differential refinement that the oxygen vibrations are appreciably greater than those of sulphur and nitrogen. Evidently, rotational-oscillation corrections to the co-ordinates of the imidodisulphate ion are needed. Accordingly, the atomic U_{ij} were analysed on the assumption that the ion is undergoing anisotropic rigid-body translations and librations (Cruickshank, 1956). In this ion, the rigid-body hypothesis is slightly unrealistic in that significant torsional oscillations of the SO_3 groups probably occur about the S–N bonds. Such internal vibrations also necessitate rotational corrections to the co-ordinates (Cruickshank, 1961*a*). However, as the corrections would be rather similar to those arising from rigid-body librations, no attempt was made to apportion the total angular vibrations between the two kinds.

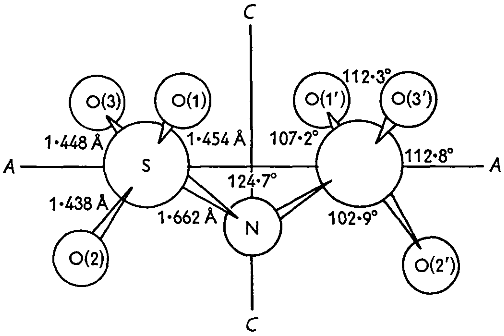


Fig. 1. Rotational axes and corrected dimensions of the imidodisulphate ion.

The molecular vibration analysis was carried out with respect to a set of standard orthogonal axes related to the original monoclinic axes by the co-ordinate transformations:

$$x' = x + z \cos\beta\,;\ y' = y\,;\ z' = z \sin\beta\ .$$

In terms of these co-ordinates, the mass centre of the ion is at (6·104, 1·355, 5·380 Å), which is within 0·025 Å of the mid-point of the S–S vectors, and the direction cosines of the principal axes of inertia are

A	−0·844,	0·000,	−0·536
B	0·536,	0·000,	−0·844
C	0·000,	1·000,	0·000 .

Two of these axes are shown in Fig. 1. *A* is the axis of minimum inertia ($I_A = 304 \times 10^{-40}$ g.cm^2) and almost coincides with the S–S′ vector; axis *B* ($I_B = 936$) is perpendicular to the SNS′ plane; and axis *C* ($I_C = 940$) is the twofold-symmetry axis. Within experimental error, the principal axes of both the translational and rotational vibration tensors coincide with the principal axes of inertia. The indicated r.m.s. amplitudes of translational vibration of the ion are 0·18 Å parallel

Table 3. *Observed and calculated structure factors*
($\times 10$)

In the $|F_o|$ list, entries in brackets are the maximum values for unobserved reflexions
All $|F_o|$ values are 1·064 times those used by Jeffrey & Jones (1956)

h	k	l	$10\lvert F_o\rvert$	$10\ F_c$
0	0	2	509	−454
0	0	4	934	−832
0	0	6	422	357
0	0	8	467	446
0	2	2	180	143
0	2	3	939	815
0	2	4	94	35
0	2	5	891	−827
0	2	6	374	−345
0	2	7	(73)	−63
0	2	8	136	−170
0	2	9	336	335
0	4	0	377	−371
0	4	1	240	−243
0	4	2	377	349
0	4	3	517	−432
0	4	4	584	556
0	4	5	684	659
0	4	6	228	−200
0	4	7	105	−86
0	4	8	129	−148
0	6	0	563	562
0	6	1	183	−167
0	6	2	178	−159
0	6	3	121	109
0	6	4	454	−499
0	6	5	94	83
0	6	6	162	204
0	6	7	(50)	19
0	8	0	145	−140
0	8	1	257	264
0	8	2	132	103
0	8	3	162	135
0	8	4	(40)	7
0	8	5	170	−189
1	1	0	223	−266
1	1	1	417	−456
1	1	2	(46)	−5
1	1	3	619	623
1	1	4	217	−218
1	1	5	23	19
1	1	6	68	30
1	1	7	189	168
1	1	8	94	77
1	1	9	(60)	−34
1	3	0	340	−367
1	3	1	33	−26
1	3	2	305	−321
1	3	3	186	184
1	3	4	313	−310
1	3	5	276	−271
1	3	6	159	136
1	3	7	279	−237
1	3	8	(40)	5
1	5	0	157	−168
1	5	1	185	197
1	5	2	474	−451
1	5	3	119	−122
1	5	4	553	543
1	5	5	51	−28
1	5	6	43	38
1	5	7	105	105
1	7	0	117	123
1	7	1	76	−71
1	7	2	160	143
1	7	3	366	−319
1	7	4	250	−243
1	7	5	(33)	12
1	7	6	60	−53
1	9	0	189	158
1	9	1	112	98
1	9	2	78	74
1	9	3	214	176
$\bar{1}$	1	1	503	−627
$\bar{1}$	1	2	89	115
$\bar{1}$	1	3	148	−157
$\bar{1}$	1	4	237	224
$\bar{1}$	1	5	137	158
$\bar{1}$	1	6	197	207
$\bar{1}$	1	7	123	115
$\bar{1}$	1	8	125	−115
$\bar{1}$	3	1	316	383
$\bar{1}$	3	2	348	330
$\bar{1}$	3	3	134	−144
$\bar{1}$	3	4	191	−168
$\bar{1}$	3	5	347	−323
$\bar{1}$	3	6	(52)	−26
$\bar{1}$	3	7	191	168
$\bar{1}$	3	8	72	73
$\bar{1}$	5	1	384	−416
$\bar{1}$	5	2	406	410
$\bar{1}$	5	3	194	192
$\bar{1}$	5	4	385	376
$\bar{1}$	5	5	60	67
$\bar{1}$	5	6	218	−242
$\bar{1}$	5	7	23	−27
$\bar{1}$	7	1	447	−444
$\bar{1}$	7	2	340	−335
$\bar{1}$	7	3	(68)	−15
$\bar{1}$	7	4	(40)	−39
$\bar{1}$	7	5	246	262
$\bar{1}$	9	1	283	286
$\bar{1}$	9	2	102	−75
$\bar{1}$	9	3	145	−117
2	0	0	134	131
2	0	4	76	70
2	0	6	778	−724
2	0	8	396	386
2	2	0	373	−423
2	2	1	938	−1086
2	2	2	780	−760
2	2	3	1018	969
2	2	4	380	375
2	2	5	364	360
2	2	6	258	246
2	2	7	456	−472
2	2	8	155	−200
2	4	0	162	171
2	4	1	247	239
2	4	2	461	−460
2	4	3	964	−882
2	4	4	229	190
2	4	5	(46)	18
2	4	6	323	337
2	4	7	279	290
2	4	8	136	−143
2	6	0	105	−113
2	6	1	189	188
2	6	2	572	560
2	6	3	39	−32
2	6	4	210	−231
2	6	5	65	−78
2	6	6	214	−218
2	6	7	107	113
2	8	0	118	−116
2	8	1	267	−300
2	8	2	(40)	−8
2	8	3	187	180
2	8	4	72	78
2	8	5	59	37
$\bar{2}$	0	4	698	670
$\bar{2}$	0	6	929	882
$\bar{2}$	0	8	293	−268
$\bar{2}$	2	1	981	1165
$\bar{2}$	2	2	367	−288
$\bar{2}$	2	3	1505	−1458
$\bar{2}$	2	4	247	−208
$\bar{2}$	2	5	126	−120
$\bar{2}$	2	6	435	−417
$\bar{2}$	2	7	405	421
$\bar{2}$	2	8	125	151
$\bar{2}$	4	1	393	−414
$\bar{2}$	4	2	729	720
$\bar{2}$	4	3	654	574
$\bar{2}$	4	4	294	−258
$\bar{2}$	4	5	104	97
$\bar{2}$	4	6	101	−95
$\bar{2}$	4	7	290	−293
$\bar{2}$	4	8	202	211
$\bar{2}$	6	1	107	97
$\bar{2}$	6	2	581	−544
$\bar{2}$	6	3	(60)	5
$\bar{2}$	6	4	58	48
$\bar{2}$	6	5	63	−82
$\bar{2}$	6	6	248	263
$\bar{2}$	6	7	132	−117
$\bar{2}$	8	1	148	145
$\bar{2}$	8	2	108	115
$\bar{2}$	8	3	139	−183
$\bar{2}$	8	4	76	82
$\bar{2}$	8	5	83	−68
3	1	0	331	−407
3	1	1	631	697
3	1	2	453	413
3	1	3	279	−299
3	1	4	93	92
3	1	5	356	350
3	1	6	69	−63
3	1	7	242	256
3	1	8	125	−144
3	1	9	35	−53
3	3	0	794	937
3	3	1	846	−938
3	3	2	205	208
3	3	3	150	128
3	3	4	351	−386
3	3	5	85	73
3	3	6	168	−163
3	3	7	102	−93
3	3	8	115	134
3	5	0	260	279
3	5	1	303	316
3	5	2	264	−249
3	5	3	504	458
3	5	4	136	−118
3	5	5	116	−96
3	5	6	207	206
3	5	7	33	−36
3	7	0	163	−183
3	7	1	318	328
3	7	2	(52)	13
3	7	3	162	−149
3	7	4	198	206
3	7	5	102	−130
3	7	6	162	−166
3	9	0	40	−33
3	9	1	218	−220
3	9	2	126	116
$\bar{3}$	1	1	636	−780
$\bar{3}$	1	2	657	726
$\bar{3}$	1	3	199	−180
$\bar{3}$	1	4	59	−40
$\bar{3}$	1	5	401	387
$\bar{3}$	1	6	222	−196
$\bar{3}$	1	7	304	−293
$\bar{3}$	1	8	77	−66
$\bar{3}$	3	1	143	−134
$\bar{3}$	3	2	622	−703
$\bar{3}$	3	3	194	138
$\bar{3}$	3	4	168	−156
$\bar{3}$	3	5	119	104
$\bar{3}$	3	6	227	209
$\bar{3}$	3	7	185	168
$\bar{3}$	3	8	51	−51
$\bar{3}$	5	1	345	346
$\bar{3}$	5	2	513	500
$\bar{3}$	5	3	107	97
$\bar{3}$	5	4	363	−342
$\bar{3}$	5	5	284	−289
$\bar{3}$	5	6	102	−105
$\bar{3}$	5	7	105	94
$\bar{3}$	7	1	(56)	27
$\bar{3}$	7	2	142	−141
$\bar{3}$	7	3	367	323
$\bar{3}$	7	4	261	266
$\bar{3}$	7	5	23	41
$\bar{3}$	7	6	136	−141
$\bar{3}$	9	1	53	−46
$\bar{3}$	9	2	128	−120
$\bar{3}$	9	3	177	−154
4	0	2	105	−92
4	0	4	706	637
4	0	6	333	−294
4	0	8	335	−285
4	2	0	724	706
4	2	1	232	−268
4	2	2	867	−807
4	2	3	612	−754
4	2	4	139	−121
4	2	5	717	685
4	2	6	213	222
4	2	7	(56)	30
4	2	8	134	180
4	4	0	401	433
4	4	1	494	480
4	4	2	85	95
4	4	3	76	79
4	4	4	532	−546
4	4	5	579	−587
4	4	6	80	63
4	4	7	51	35
4	4	8	105	144
4	6	0	553	−575
4	6	1	137	133
4	6	2	212	210
4	6	3	180	179
4	6	4	253	268
4	6	5	51	48
4	6	6	148	−126
4	8	0	81	85
4	8	1	189	−192
4	8	2	241	−227
4	8	3	126	−118
4	8	4	(34)	19
$\bar{4}$	0	2	868	883
$\bar{4}$	0	4	1098	1161
$\bar{4}$	0	6	320	−306
$\bar{4}$	0	8	203	−193
$\bar{4}$	2	1	986	−1006
$\bar{4}$	2	2	176	−145
$\bar{4}$	2	3	630	−539
$\bar{4}$	2	4	218	−201
$\bar{4}$	2	5	711	683
$\bar{4}$	2	6	(65)	−38
$\bar{4}$	2	7	49	−48
$\bar{4}$	2	8	(60)	72
$\bar{4}$	4	1	296	261
$\bar{4}$	4	2	168	−143
$\bar{4}$	4	3	489	407
$\bar{4}$	4	4	162	−153
$\bar{4}$	4	5	231	−210
$\bar{4}$	4	6	332	319
$\bar{4}$	4	7	45	−53
$\bar{4}$	4	8	67	70
$\bar{4}$	6	1	191	176
$\bar{4}$	6	2	117	103
$\bar{4}$	6	3	364	−303
$\bar{4}$	6	4	243	229
$\bar{4}$	6	5	72	−97
$\bar{4}$	6	6	119	−106
$\bar{4}$	8	1	242	−243
$\bar{4}$	8	2	208	186
$\bar{4}$	8	3	(53)	10
$\bar{4}$	8	4	83	−92
5	1	0	41	−35
5	1	1	972	1224
5	1	2	248	−265
5	1	3	73	−63
5	1	4	(40)	−34
5	1	5	310	−292
5	1	6	366	351
5	1	7	183	184
5	1	8	70	−76
5	3	0	672	−738
5	3	1	343	−346
5	3	2	702	670
5	3	3	367	−339
5	3	4	432	436
5	3	5	133	−116
5	3	6	363	−362
5	3	7	159	164
5	3	8	28	−6
5	5	0	308	307
5	5	1	541	−571
5	5	2	91	67
5	5	3	248	230
5	5	4	237	−237
5	5	5	230	258
5	5	6	36	33
5	5	7	115	−89
5	7	0	72	71
5	7	1	123	120
5	7	2	258	−259
5	7	3	53	44
5	7	4	125	134
5	7	5	90	−110
5	9	0	103	−114
5	9	1	(23)	6
$\bar{5}$	1	1	250	−278
$\bar{5}$	1	2	211	241
$\bar{5}$	1	3	601	596
$\bar{5}$	1	4	515	−477
$\bar{5}$	1	5	620	−598
$\bar{5}$	1	6	(73)	4
$\bar{5}$	1	7	248	−246
$\bar{5}$	1	8	199	199
$\bar{5}$	3	1	97	−91
$\bar{5}$	3	2	530	−513
$\bar{5}$	3	3	331	295
$\bar{5}$	3	4	600	548
$\bar{5}$	3	5	167	158
$\bar{5}$	3	6	117	103
$\bar{5}$	3	7	72	−62
$\bar{5}$	3	8	264	−304
$\bar{5}$	5	1	110	−103
$\bar{5}$	5	2	327	−325
$\bar{5}$	5	3	506	−533
$\bar{5}$	5	4	169	−159
$\bar{5}$	5	5	23	20
$\bar{5}$	5	6	112	134
$\bar{5}$	5	7	152	186
$\bar{5}$	7	1	305	313
$\bar{5}$	7	2	218	214
$\bar{5}$	7	3	102	92
$\bar{5}$	7	4	80	−96
$\bar{5}$	9	1	102	−136
6	0	0	714	711
6	0	2	1071	−1065
6	0	4	103	75
6	0	6	148	135
6	0	8	260	−230
6	2	0	473	460
6	2	1	631	659
6	2	2	587	585
6	2	3	337	−321
6	2	4	183	−171
6	2	5	110	−111
6	2	6	94	−93
6	2	7	202	232
6	2	8	136	178
6	4	0	233	−238
6	4	1	(40)	40
6	4	2	269	286

Table 3 (*cont.*)

h	k	l	10|F_o|	10 F_c
6	4	3	332	353
6	4	4	(35)	9
6	4	5	(37)	-11
6	4	6	272	-295
6	4	7	214	-248
6	6	0	74	-65
6	6	1	243	-265
6	6	2	180	-178
6	6	3	(60)	20
6	6	4	117	142
6	6	5	222	259
6	6	6	138	165
6	8	0	209	200
6	8	1	109	121
6	8	2	33	23
6	8	3	88	-81
$\bar{6}$	0	2	351	320
$\bar{6}$	0	4	179	-159
$\bar{6}$	0	6	541	-479
$\bar{6}$	0	8	274	243
$\bar{6}$	2	1	267	-261
$\bar{6}$	2	2	58	-75
$\bar{6}$	2	3	637	589
$\bar{6}$	2	4	223	-205
$\bar{6}$	2	5	31	-19
$\bar{6}$	2	6	127	136
$\bar{6}$	2	7	205	-272
$\bar{6}$	2	8	38	-41
$\bar{6}$	4	1	652	742
$\bar{6}$	4	2	252	-247
$\bar{6}$	4	3	257	-220
$\bar{6}$	4	4	274	258
$\bar{6}$	4	5	(40)	-27
$\bar{6}$	4	6	(41)	26
$\bar{6}$	4	7	85	78
$\bar{6}$	6	1	274	-282
$\bar{6}$	6	2	314	301
$\bar{6}$	6	3	94	-96
$\bar{6}$	6	4	50	-64
$\bar{6}$	6	5	129	136
$\bar{6}$	6	6	157	-144
$\bar{6}$	8	1	(42)	24
$\bar{6}$	8	2	33	-35
$\bar{6}$	8	3	174	160
$\bar{6}$	8	4	198	-206
7	1	0	290	334
7	1	1	77	-90
7	1	2	84	75
7	1	3	1023	968
7	1	4	492	-495
7	1	5	340	-318
7	1	6	60	-14
7	1	7	223	-257
7	1	8	170	185
7	3	0	668	-718
7	3	1	169	168
7	3	2	402	-430
7	3	3	36	-13
7	3	4	440	450
7	3	5	103	-140
7	3	6	195	203
7	3	7	41	-12
7	5	0	69	68
7	5	1	(56)	17
7	5	2	29	-25
7	5	3	397	-393
7	5	4	(27)	-23
7	5	5	97	87
7	5	6	100	-152
7	7	0	211	212
7	7	1	94	-104
7	7	2	87	86
7	7	3	203	183
7	7	4	107	-146
$\bar{7}$	1	1	405	462
$\bar{7}$	1	2	327	-321
$\bar{7}$	1	3	310	-317
$\bar{7}$	1	4	100	-102
$\bar{7}$	1	5	627	-656
$\bar{7}$	1	6	315	327
$\bar{7}$	1	7	262	284
$\bar{7}$	1	8	(47)	-56
$\bar{7}$	3	1	165	176
$\bar{7}$	3	2	917	1013
$\bar{7}$	3	3	198	192
$\bar{7}$	3	4	233	209
$\bar{7}$	3	5	(34)	-14
$\bar{7}$	3	6	498	-426
$\bar{7}$	3	7	71	66
$\bar{7}$	5	1	648	-705
$\bar{7}$	5	2	373	-375
$\bar{7}$	5	3	103	96
$\bar{7}$	5	4	171	165
$\bar{7}$	5	5	262	284
$\bar{7}$	5	6	62	68
$\bar{7}$	7	1	119	131
$\bar{7}$	7	2	103	-96
$\bar{7}$	7	3	171	-139
$\bar{7}$	7	4	69	-79
8	0	0	164	135
8	0	2	344	-315
8	0	4	113	-89
8	0	6	(66)	-17
8	2	0	83	-94
8	2	1	(39)	31
8	2	2	444	433
8	2	3	169	173
8	2	4	209	214
8	2	5	143	-147
8	2	6	(62)	-30
8	2	7	(52)	-24
8	4	0	252	-279
8	4	1	239	-255
8	4	2	266	-267
8	4	3	87	-79
8	4	4	151	145
8	4	5	256	271
8	4	6	36	52
8	4	7	84	-111
8	6	0	140	141
8	6	1	134	118
8	6	2	41	-54
8	6	3	265	-246
8	6	4	(48)	-51
8	6	5	64	-78
8	8	0	86	-83
8	8	1	(33)	-29
8	8	2	261	277
$\bar{8}$	0	2	583	-551
$\bar{8}$	0	4	295	-244
$\bar{8}$	0	6	77	56
$\bar{8}$	0	8	67	82
$\bar{8}$	2	1	276	294
$\bar{8}$	2	2	195	-193
$\bar{8}$	2	3	102	-94
$\bar{8}$	2	4	259	249
$\bar{8}$	2	5	136	-136
$\bar{8}$	2	6	85	95
$\bar{8}$	2	7	(48)	-26
$\bar{8}$	4	1	145	128
$\bar{8}$	4	2	165	155
$\bar{8}$	4	3	367	-315
$\bar{8}$	4	4	118	-85
$\bar{8}$	4	5	(33)	36
$\bar{8}$	4	6	118	-114
$\bar{8}$	4	7	98	120
$\bar{8}$	6	1	189	-206
$\bar{8}$	6	2	103	95
$\bar{8}$	6	3	217	181
$\bar{8}$	6	4	92	-106
$\bar{8}$	6	5	37	43
$\bar{8}$	8	1	51	58
$\bar{8}$	8	2	198	-206
9	1	0	312	-339
9	1	1	633	-778
9	1	2	473	505
9	1	3	112	122
9	1	4	70	83
9	1	5	387	385
9	1	6	335	-288
9	1	7	223	-192
9	3	0	742	779
9	3	1	144	-155
9	3	2	436	-435
9	3	3	173	177
9	3	4	240	-266
9	3	5	90	80
9	3	6	346	369
9	3	7	(33)	11
9	5	0	157	-163
9	5	1	409	462
9	5	2	80	74
9	5	3	51	-44
9	5	4	33	45
9	5	5	281	-270
9	7	0	46	-41
9	7	1	163	-176
9	7	2	127	123
9	7	3	103	74
$\bar{9}$	1	1	82	-91
$\bar{9}$	1	2	154	-144
$\bar{9}$	1	3	706	-720
$\bar{9}$	1	4	228	210
$\bar{9}$	1	5	285	315
$\bar{9}$	1	6	83	25
$\bar{9}$	1	7	179	180
$\bar{9}$	3	1	(46)	-28
$\bar{9}$	3	2	367	336
$\bar{9}$	3	3	117	103
$\bar{9}$	3	4	562	-535
$\bar{9}$	3	5	(32)	-26
$\bar{9}$	3	6	71	-54
$\bar{9}$	3	7	93	103
$\bar{9}$	5	1	118	123
$\bar{9}$	5	2	57	-53
$\bar{9}$	5	3	546	420
$\bar{9}$	5	4	51	64
$\bar{9}$	5	5	216	-231
$\bar{9}$	7	1	(34)	-12
$\bar{9}$	7	2	125	-118
$\bar{9}$	7	3	161	-123
10	0	0	256	-220
10	0	2	214	-188
10	0	4	156	-152
10	2	0	48	42
10	2	1	87	96
10	2	2	108	-111
10	2	3	104	-126
10	2	4	172	192
10	2	5	42	32
10	2	6	(52)	-30
10	4	0	86	72
10	4	1	98	-104
10	4	2	85	-74
10	4	3	70	-71
10	4	4	162	-220
10	4	5	84	90
10	4	6	58	70
10	6	0	75	73
10	6	1	222	262
10	6	2	(41)	38
10	6	3	116	58
$\overline{10}$	0	2	98	73
$\overline{10}$	0	4	122	-102
$\overline{10}$	0	6	72	-65
$\overline{10}$	2	1	(56)	14
$\overline{10}$	2	2	88	94
$\overline{10}$	2	3	60	-61
$\overline{10}$	2	4	348	353
$\overline{10}$	2	5	102	116
$\overline{10}$	2	6	(49)	-46
$\overline{10}$	2	7	33	-55
$\overline{10}$	4	1	202	-276
$\overline{10}$	4	2	77	-58
$\overline{10}$	4	3	105	-92
$\overline{10}$	4	4	169	-168
$\overline{10}$	4	5	136	146
$\overline{10}$	4	6	65	62
$\overline{10}$	6	1	245	248
$\overline{10}$	6	2	94	81
$\overline{10}$	6	3	(53)	53
$\overline{10}$	6	4	65	-95
11	1	0	365	-388
11	1	1	(56)	-46
11	1	2	156	-154
11	1	3	627	-627
11	1	4	235	255
11	1	5	159	137
11	1	6	152	119
11	3	0	390	375
11	3	1	46	-13
11	3	2	329	318
11	3	3	43	20
11	3	5	37	-21
11	3	6	56	-81
11	5	0	187	-186
11	5	1	125	-128
11	5	2	(23)	-14
11	5	3	305	264
11	5	4	103	105
$\overline{11}$	1	1	419	-487
$\overline{11}$	1	2	274	292
$\overline{11}$	1	3	223	219
$\overline{11}$	1	4	(35)	16
$\overline{11}$	1	5	266	324
$\overline{11}$	1	6	174	-164
$\overline{11}$	3	1	148	-151
$\overline{11}$	3	2	553	-551
$\overline{11}$	3	3	(46)	44
$\overline{11}$	3	4	107	-92
$\overline{11}$	3	5	23	-29
$\overline{11}$	3	6	314	371
$\overline{11}$	5	1	310	336
$\overline{11}$	5	2	46	48
$\overline{11}$	5	3	150	-143
$\overline{11}$	5	4	49	51
12	0	0	350	324
12	0	2	(52)	32
12	0	4	192	-163
12	0	6	(47)	0
12	2	0	54	42
12	2	1	145	156
12	2	2	60	-61
12	2	3	(60)	36
12	2	4	(56)	87
12	2	5	(49)	-64
12	4	0	89	-83
12	4	1	81	84
12	4	2	159	141
12	4	3	115	-97
12	4	4	(29)	19
12	6	0	85	69
12	6	1	155	-161
12	6	2	98	105
$\overline{12}$	0	2	239	-228
$\overline{12}$	0	4	211	-178
$\overline{12}$	0	6	(36)	9
$\overline{12}$	2	1	208	221
$\overline{12}$	2	2	210	208
$\overline{12}$	2	3	89	101
$\overline{12}$	2	4	(32)	14
$\overline{12}$	2	5	72	-120
$\overline{12}$	4	1	129	-135
$\overline{12}$	4	2	129	-128
$\overline{12}$	4	3	(36)	45
$\overline{12}$	4	4	99	70
$\overline{12}$	6	1	127	133
$\overline{12}$	6	2	48	-50
13	1	0	189	187
13	1	1	268	267
13	1	2	220	-204
13	1	3	98	-91
13	1	4	185	-156
13	1	5	380	-293
13	3	0	343	-339
13	3	1	109	119
13	3	2	261	260
13	3	3	125	103
13	3	4	67	86
13	5	0	60	63
13	5	1	257	-255
13	5	2	190	-190
$\overline{13}$	1	1	63	42
$\overline{13}$	1	2	159	151
$\overline{13}$	1	3	538	441
$\overline{13}$	1	4	160	-168
$\overline{13}$	1	5	125	-148
$\overline{13}$	3	1	(43)	13
$\overline{13}$	3	2	103	-96
$\overline{13}$	3	3	50	-33
$\overline{13}$	3	4	278	277
$\overline{13}$	5	1	107	-104
$\overline{13}$	5	2	77	81
14	0	0	51	-36
14	0	2	267	223
14	0	4	117	90
14	2	0	(46)	25
14	2	1	194	-207
14	2	2	89	-97
14	2	3	145	138
14	4	0	78	-75
14	4	1	197	217
14	4	2	83	-86
$\overline{14}$	0	2	451	-371
$\overline{14}$	0	4	197	198
$\overline{14}$	2	1	125	131
$\overline{14}$	2	2	56	57
$\overline{14}$	2	3	265	-213
$\overline{14}$	2	4	94	-102
$\overline{14}$	4	1	(33)	36
$\overline{14}$	4	2	121	111
15	1	0	115	99
15	1	1	33	-29
15	1	2	157	147
15	1	3	(244)	192
15	3	0	130	-144
15	3	1	71	-80
$\overline{15}$	1	1	214	253
$\overline{15}$	1	2	145	-99
$\overline{15}$	1	3	40	-4
$\overline{15}$	3	1	36	-37
$\overline{15}$	3	2	119	161
16	0	0	290	-243
0	2	0	973	-1148
0	2	1	1233	1420
2	0	2	1714	2204
$\bar{2}$	0	2	1597	-1874
4	0	0	1336	-1886
$\bar{4}$	6	7	38	110
5	7	5	136	-242
10	6	4	19	-85
11	3	4	240	-282

Table 4. *Observed and calculated vibration tensor components with respect to inertial axes*

(All values in Å^2)

Atom	U_{11} obs.	U_{11} calc.	U_{22} obs.	U_{22} calc.	U_{33} obs.	U_{33} calc.	U_{12} obs.	U_{12} calc.	U_{13} obs.	U_{13} calc.	U_{23} obs.	U_{23} calc.
S	0·0300	0·0310	0·0262	0·0269	0·0323	0·0321	0·0003	0·0001	−0·0014	0·0002	0·0001	−0·0001
O(1)	0·0420	0·0391	0·0385	0·0345	0·0600	0·0615	0·0010	0·0072	−0·0033	−0·0058	0·0175	0·0133
O(2)	0·0392	0·0364	0·0547	0·0576	0·0455	0·0471	−0·0049	−0·0030	0·0058	0·0113	−0·0011	−0·0012
O(3)	0·0460	0·0392	0·0469	0·0377	0·0437	0·0451	−0·0025	−0·0072	−0·0116	−0·0043	−0·0057	−0·0120
N	0·0414	0·0337	0·0289	0·0271	0·0280	0·0227	−0·0008	−0·0007	0	0	0	0

to *A*, 0·14 Å parallel to *B*, and 0·15 Å parallel to *C*; the apparent r.m.s. angular oscillations are 6·8° about *A*, 3·7° about *B*, and 3·9° about *C*. Thus the axis associated with the largest oscillations is that with the minimum inertia and the least cross-section. The same effect was found in the $K_2S_2O_7$ and $K_2[CH_2(SO_3)_2]$ structures, though in the former the apparent translation vibrations were rather smaller than in the present structure. This difference is probably due to a slight systematic error in one or other set of experimental data, most likely the neglect of absorption corrections for the pyrosulphate. However, this possible systematic error is not important for the present purpose, since the chief aim of the vibration analysis is to obtain the rotational corrections for the co-ordinates. The U_{ij} calculated with the above translational and librational rigid-body vibrations together with the observed U_{ij} with respect to the inertial axes are shown in Table 4; there is an r.m.s. difference of 0·0040 Å^2 between the observed and calculated values. Since this is not much greater than the 0·0030 Å^2 estimated earlier for the oxygen U_{ii} e.s.d.'s, the rigid-body hypothesis evidently accounts for the major part of the anisotropic vibrations, and the derived librational amplitudes may justifiably be used to calculate the rotational corrections for the co-ordinates. Table 5 gives the revised co-ordinates; the largest correction is 0·012 Å to the *z* co-ordinate of O(1).

Table 5. *Monoclinic co-ordinates of the anion after rotational correction*

Atom	*x* (Å)	*y* (Å)	*z* (Å)
S	4·9530	1·3294	4·5864
O(1)	5·5151	2·0314	3·4460
O(2)	4·0725	0·2418	4·2215
O(3)	4·3665	2·2432	5·5455
N	6·2150 (= *a*/2)	0·5677	5·3812 (= 3*c*/4)
H	6·2150 (= *a*/2)	−0·4400	5·3812 (= 3*c*/4)

Discussion

Table 6 gives the bond lengths and angles obtained from both the uncorrected and corrected co-ordinates. (See Fig. 1 for the numbering of the atoms.) The corrected dimensions differ by at most 0·009 Å and 0·8° from those given by Jeffrey & Jones, whose work is thus satisfyingly corroborated by the present more detailed refinement. From the co-ordinate e.s.d.'s given above, the e.s.d.'s in the individual dimensions are calculated as S–O 0·006 Å, S–N 0·004 Å, ∠ O–S–O and ∠ N–S–O 0·3°, ∠ S–N–S 0·5°. The three S–O bond lengths are equal within experimental error and their mean is 0·006 Å greater than the Jeffrey & Jones mean, chiefly because of the rotational corrections. The e.s.d. of the mean is roughly $0·006/\sqrt{3} = 0·004$ Å, but to allow for the effects of some neglected off-diagonal terms in the least-squares matrix and for any inaccuracies in the rotational corrections, it would be prudent to increase the e.s.d.'s of S–O (mean) and S–N from 0·004 to 0·005 Å. The mean S–O length is therefore 1·453 ± 0·005 Å and S–N is 1·662 ± 0·005 Å. The variations among the individual O–S–O angles and among the O–S–N angles are just outside experimental error and presumably are due to steric effects. The mean O–S–O angle is 113·0° and the mean O–S–N angle is 105·6. The revised values for the shorter intramolecular distances between non-bonded atoms are shown also in Table 6; there are no important differences from the values given by Jeffrey & Jones.

Table 6. *Molecular dimensions*

		Jeffrey & Jones	Present uncorrected	Rotationally corrected
	S–N	1·655 Å	1·654 Å	1·662 Å
	S–O(1)	1·454	1·451	1·461
	S–O(2)	1·438	1·434	1·442
	S–O(3)	1·448	1·446	1·457
Mean	S–O	1·447	1·444	1·453
	S–N–S	124·7°	125·7°	125·5°
	O(1)–S–O(2)	113·5	114·0	114·0
	O(2)–S–O(3)	112·8	112·7	112·8
	O(3)–S–O(1)	112·3	112·3	112·3
	N–S–O(1)	107·2	107·0	107·0
	N–S–O(2)	102·9	103·5	103·2
	N–S–O(3)	106·9	106·5	106·5

Corrected intramolecular distances between non-bonded atoms:

O(1)–O(2)	2·44 Å	O(2)–O(3)	2·41 Å	O(3)–O(1)	2·42 Å
N–O(1)	2·51	N–O(2)	2·44	N–O(3)	2·50
O(1)–O(3′)	3·08	S–S′	2·96		

Jeffrey & Jones reported that they had been unable to locate the hydrogen electron density unequivocally by difference synthesis, and that the planar configuration, rather than a statistical pyramidal one, about the nitrogen atom could be inferred only from the S–N bond lengths. We have been no more successful with the least-squares technique, as the inclusion of the hydrogen atom, whether on the axis or in a statistical distribution of half-atoms off the axis, gave no improvement to the residual and led to implausible parameter shifts for the atom. Siebert (1957) reports a single infra-red frequency centred on 3235 cm^{-1} for the NH stretching vibration, but it is rather broad and thus leaves the nature of the H sites in doubt. However, in the electron spin resonance spectrum of an irradiated potassium imidodisulphate crystal, Horsfield, Morton, Rowlands & Whiffen (1962) detected at certain orientations a small splitting of the ^{14}N triplet in the trapped $^{\cdot}\mathrm{N(SO_3)_2^{=}}$ radical; this is attributed to interactions with hydrogen nuclei in the host imidodisulphate ions. On the whole, the e.s.r. line shape tends to favour the existence of a single hydrogen site in the normal molecule.

In his theoretical discussion of π-bonds involving the $d_{x^2-y^2}$ and d_{z^2} orbitals of sulphur, Cruickshank (1961*b*) has carried considerably further the interpetation of the dimensions of the imidodisulphate ion given by Jeffrey & Jones. Here, we remark only that the S–N bond of 1·662 ± 0·005 Å is of a similar length to the S–O(4) (bridge) bond of 1·645 ± 0·005 Å in the pyrosulphate ion (Lynton & Truter, 1960). On Cruickshank's theory, they are both partial double bonds in contrast to the appreciably larger S–C bond of 1·770 ± 0·007 Å in the methylenedisulphate (Truter, 1962), which cannot easily acquire any double-bond character since the carbon is bonded to four other atoms.

The slight differences of about 0·2 Å in the cell dimensions of the isostructural compounds $K_2S_2O_7$, $K_2[NH(SO_3)_2]$, and $K_2[CH_2(SO_3)_2]$ may be attributed partly to the extra space required for the hydrogen atom in the imidodisulphate and for the two hydrogen atoms in the methylenedisulphate. A second cause is that the intra-ionic dimensions of the common parts of the anions also differ by as much as 0·1 Å.

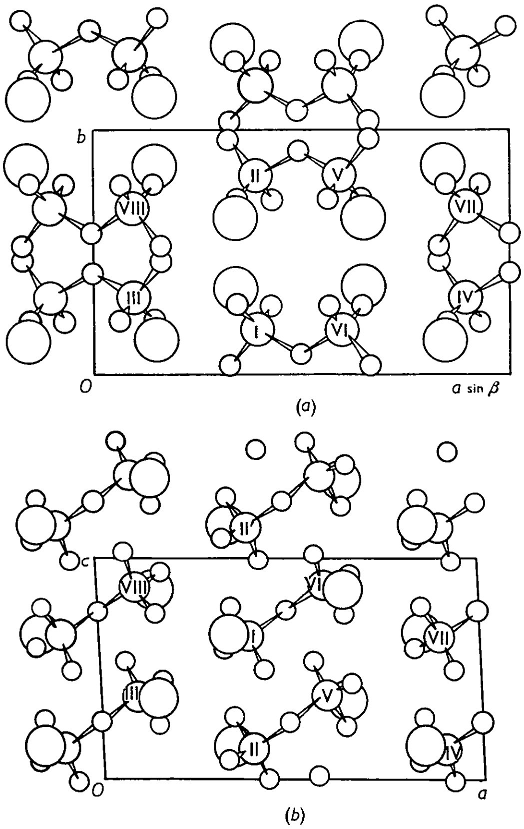
Table 7. *The shorter* K · · · O *interatomic distances*

(All values in Å)

K · · · O(1)	3·23(I)	2·75(II)	2·83(V)
K · · · O(2)	2·95(I′)	3·00(III)	3·05(VIII)
K · · · O(3)	2·71(I)	2·70(II)	2·82(VIII)

These distances refer to the cation K(I), and the roman numerals are those on the anions in Fig. 2(*a*).

In each crystal structure, the principal interionic contacts are between potassium and oxygen atoms; for potassium imidodisulphate, the revised K · · · O distances are given in Table 7. In all three structures, these sets of K · · · O contacts are very similar. On the other hand, there are important differences in the way contacts are made with the bridging groups: O(4), NH, and CH_2. For convenience, Jeffrey & Jones's illustrations of the $K_2[NH(SO_3)_2]$ structure are reproduced as Fig. 2; the corresponding illustrations of the other two structures would be sufficiently similar.



(*a*)

(*b*)

Fig. 2. Crystal structure of $K_2[NH(SO_3)_2]$ projected (*a*) down [001] and (*b*) along [010].

In the pyrosulphate, the bridge atom O(4) is not involved in any electrostatic or interionic van der Waals contacts, as the O(4) of group I/VI is 3·31 Å from the O(1)'s and 3·36 Å from the O(2)'s of the groups II′ and V. [See Fig. 2(*b*).] In the imidodisulphate, though the nitrogen is at similar distances 3·17 and 3·41 Å, from the O(1)'s and O(2)'s, the hydrogen (assumed coplanar with the nitrogen and sulphur atoms) is only 2·41 Å from the two O(1)'s; this distance is appropriate to a van der Waals contact. In the methylenedisulphate, the hydrogens are no longer on the twofold axis and each is close to an O(1) (2·54 Å), an O(2) (2·43 Å), an S (2·74 Å) and an H (2·34 Å) of the adjacent group (the carbon is 3·47 Å from two O(1)'s and 3·39 Å from two O(2)'s).

While most of the interionic O · · · O distances in this structure are in excess of 3·2 Å, there is in each structure one interionic O · · · O separation which is only slightly longer than a van der Waals distance. This is the contact from O(3) of I to O(2) of VIII; the distances are 3·04, 3·00, and 3·13 Å in the three salts of pyrosulphate, imidodisulphate, and methylenedisulphate respectively.

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The Crystal Structure of Trimethyloxosulfonium Fluoborate $[(CH_3)_3SO]^+BF_4^-$

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Crystals of $[(CH_3)_3SO]^+BF_4^-$ are orthorhombic with unit cell dimensions

$$a = 11{\cdot}49 \pm 0{\cdot}01,\ b = 11{\cdot}80 \pm 0{\cdot}01,\ c = 11{\cdot}58 \pm 0{\cdot}01\ \text{Å}\ .$$

The space group is *Pbcn* with $Z = 8$. Although the $[(CH_3)_3SO]^+$ ion is not required crystallographically to have any symmetry, it approximates closely to symmetry $3m$. The fluoborate ions are crystallographically of two types and both are required to have the symmetry 2. However, these ions achieve this symmetry in a statistical (disordered) manner.

Bond distances and angles in the trimethyloxosulfonium ion are: S–C(1) = 1·77 ± 0·02 Å, S–C(2) = 1·78 ± 0·02 Å, S–C(3) = 1·78 ± 0·02 Å, S–O = 1·45 ± 0·01 Å, C(1)–S–C(2) = 106·8 ± 0·8°, C(1)–S–C(3) = 107·2 ± 0·8°, C(2)–S–C(3) = 105·1 ± 0·8°, C(1)–S–O = 112·6 ± 0·7°, C(2)–S–O = 112·1 ± 0·7°, C(3)–S–O = 112·7 ± 0·7°.

Because of the disorder, it was not possible to obtain accurate bond distances and angles in the fluoborate ions.

Introduction

The crystal structure of trimethyloxosulfonium perchlorate has been reported by Coulter, Gantzel & McCullough (1961, 1963). At the time it became apparent that the perchlorate is disordered, the study of the fluoborate salt was undertaken with the hope that it would be ordered and thus permit a more accurate determination of the distances and angles in the $(CH_3)_3SO^+$ ion. Although it was found that the fluoborate salt was even more disordered than the perchlorate, the distances and angles in the $(CH_3)_3SO^+$ ion are in excellent agreement with the values found in the perchlorate and the structure reported for this interesting ion in the previous paper is confirmed.

Experimental

The preparation of trimethyloxosulfonium fluoborate has been described by Smith (1959) who kindly supplied the analyzed sample used in this study. Crystals suitable for the X-ray work were grown by the slow evaporation of solutions of the salt in acetone, in which it is only slightly soluble.

Weissenberg and precession photographs about the c axis of the unit appear much like those of the perchlorate. However, on closer inspection it was apparent that the fluoborate has orthorhombic rather than tetragonal symmetry. The lattice constants are:

$$a = 11{\cdot}49 \pm 0{\cdot}01,\ b = 11{\cdot}80 \pm 0{\cdot}01,\ c = 11{\cdot}58 \pm 0{\cdot}01\ \text{Å}$$

based on Cu $K\alpha = 1{\cdot}5418$ Å. The only systematic absences are $0kl$ with k odd, $h0l$ with l odd and $hk0$ with $h+k$ odd. The space group was accordingly assumed to be *Pbcn*. The flotation density of the crystals was found to be 1·52 g.cm^{-3} while that calculated for $Z = 8$ is 1·523 g.cm^{-3}.

The intensity data were obtained from sets of multiple-film Weissenberg photographs about the b and c axes prepared with Cu $K\alpha$ radiation. The crystals were about 0·18 mm by 0·20 mm in cross-section and the corresponding value of μR for copper radia-