379. *An Addition Compound of Tetraethylammonium Bromide and Succinimide.*

By H. M. Powell and E. Wait.

A compound previously formulated in a different way has been shown to be a lattice compound containing tetraethylammonium and bromide ions and succinimide molecules. The principle of the structure is local neutralisation of charges with the positive NH group of succinimide directed towards the negative ions, the negative oxygen atoms of the succinimide towards the positive ions, and negative and positive ions themselves in close contact.

There are many crystalline substances containing molecules of more than one kind, which together build up the crystal structure as a whole without ordinary chemical linkages between the components of different sorts.[1] Intermolecular attractions between the different components are of the van der Waals type. Some related structures exist in which one of the combining substances is molecular and the other ionic. For instance, $Cs_3As_2Cl_9$ has a structure which contains $AsCl_3$ molecules with cæsium and chloride ions;[2] the compounds sucrose–sodium bromide[3] and cysteinylglycine–sodium iodide[4] are also of this type.

Braude and Waight[5] called attention to the compound under investigation, for which they had suggested a formula. It seemed likely that it might be another example of a lattice compound with $(Et_4N)Br$ as the ionic and succinimide as the molecular constituent. The preparation of this compound and of its chloride analogue by crystallisation of chloroform solutions of the appropriate components in equimolar proportions lent support to this hypothesis. Accordingly an *X*-ray crystallographic examination was undertaken.

The bromide crystals prepared as above were unsuitable for detailed *X*-ray analysis and those used were from a sample provided by Professor Braude. This was extracted from the solid residues of the reaction of *cyclo*hexene with *N*-bromosuccinimide carried out with tetraethylammonium bromide as a catalyst.[5] The chloride was deliquescent, but the bromide remained unchanged under normal atmospheric conditions. The composition of the *chloride* was established by analysis (Found: C, 53·2; H, 8·27; N, 11·1; Cl, 9·75. $C_{16}H_{30}O_4N_3Cl$ requires C, 52·9; H, 8·26; N, 11·6; Cl, 9·77%). The densities were measured by flotation and were, for the bromide 1·370 ± 0·005 g./c.c., and for the chloride 1·235 ± 0·005 g./c.c.

For the *X*-ray investigation Cu-K_α radiation was used. Oscillation and equi-inclination Weissenberg techniques were used to measure intensities of *X*-ray reflections. Unit-cell dimensions were deduced from high-angle axial reflections on Weissenberg photographs calibrated with copper-powder lines. Two crystals elongated respectively along *a* and *b* were used for the 0*kl* and *h*0*l* zones of the bromide. The chloride crystals were needles elongated along *b*. An equidimensional fragment cut from such a needle and coated with a mixture of Vaseline and liquid paraffin was used for the 0*kl* zone. Crystals could thus be preserved for about 24 hr. Relative intensities of reflections, recorded by the multiple-film technique, were measured by visual comparison with a scale of standard spots prepared with a crystal.

The bromide and chloride form similar structures. Oscillation photographs about two mutually perpendicular axes have equatorial symmetry and, with the cell dimensions and optically biaxial behaviour, establish orthorhombic symmetry. The bromide has $a = 7·52 \pm 0·02$, $b = 18·76 \pm 0·05$, $c = 27·90 \pm 0·10$ Å, and the chloride $a = 7·56 \pm 0·02$, $b = 18·78 \pm 0·05$, $c = 27·70 \pm 0·10$ Å. For eight formula units $C_{16}H_{30}O_4N_3X$

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[1] Powell, *J.*, 1954, 2658.
[2] Hoard and Goldstein, *J. Chem. Phys.*, 1935, **3**, 199.
[3] Beevers and Cochran, *Proc. Roy. Soc.*, 1947, *A*, **190**, 257.
[4] Dyer, *Acta Cryst.*, 1951, **4**, 42.
[5] Braude and Waight, *J.*, 1952, 1116.

per cell, the calculated densities are 1·379 g./c.c. for X = Br and 1·238 g./c.c. for X = Cl, in good agreement with the measured values.

The absent spectra, $0kl$, $k \neq 2n$, $h0l$, $l \neq 2n$, and $hk0$, $h \neq 2n$, show that the space-group of both compounds is *Pbca*.

The general position in the space-group is of 8-fold multiplicity and there are special 4-fold positions coinciding with symmetry centres. Patterson projections perpendicular to [100] and [010] gave the positions of the bromide atoms, showing that they do not occupy the special positions. All atoms were therefore assumed to be in general positions.

FIG. 1.

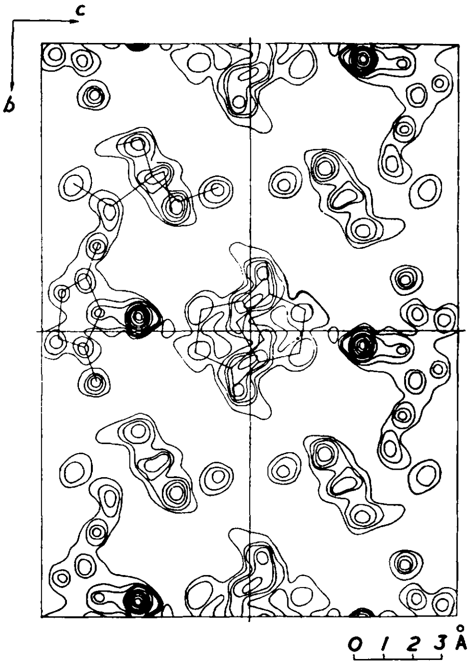

FIG. 2.

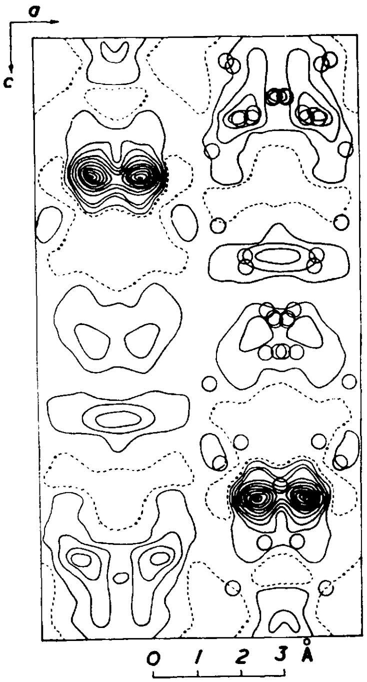

FIG. 1. *Electron density projected perpendicular to* [100]. *The origin of the unit cell is at the centre of the figure which shows the area from* $-b/2$ *to* $b/2$ *and* $-c/4$ *to* $c/4$. *Contours are at arbitrary equal intervals, approximately* 2 *electrons*/Å^2 *and at five times that interval on bromine atoms which are represented by the four high peaks. Some of the succinimide molecules and a tetraethylammonium ion are indicated in outline.*

FIG. 2. *Electron density projected perpendicular to* [010]. *The origin of the unit cell is at the centre of the figure which shows the area from* $-c/4$ *to* $c/4$ *and* $-a/2$ *to* $a/2$. *The high peaks correspond to bromine. In the right half the lighter atoms are indicated by circles.*

Approximate co-ordinates for the bromine are $x = 0{\cdot}176$, $z = 0{\cdot}135$. For y the value is 0·026 or $\frac{1}{4} - 0{\cdot}026$. The ambiguity, which is unresolvable in the two projections used, does not interfere with the refinement of the [100] projection by Fourier F synthesis. The change of the co-ordinate of the bromine atom from y to $\frac{1}{4} - y$ leaves the sign of its contribution to F_{0kl} unchanged if $k = 4n$ but reversed if $k = 4n + 2$. Fourier syntheses based on the two sets of signs appear the same if the y axis is reversed in direction and the origin is transferred to $y = \frac{1}{4}$. Similarly, when differences between F_{0kl} from bromide and chloride are considered the ambiguity has the same effect.

A Fourier synthesis based on F_{0kl} with the signs determined by bromine contributions alone did not lead directly to further development of the structure. On the assumption that the halogen parameters were sufficiently close for application of the isomorphous replacement method, a set of signs was obtained by comparison of the intensities of corresponding reflections of the bromide and chloride crystals. The absolute scale of the observed structure factors and an approximate average value for the temperature factor were determined for each compound by Wilson's method.[6] The resulting Fourier syntheses for the two compounds were very similar. Several features common to both could be interpreted as atomic groups that might be expected in the structure. These included the tetraethylammonium group and one succinimide molecule. The projection perpendicular to [010] with signs based on the heavy-atom contribution was calculated for the bromide. This showed the same succinimide molecule edge-on and indicated its slope. The position of the remaining succinimide molecule was also suggested by this projection. Three cycles of Fourier refinement of the [100] projection now followed. At the second stage an isotropic temperature factor, different from that of the light atoms, was assigned to the bromine atom. Thus the structure factor F was assumed to be of the form:

$$n\{F_{\mathrm{Br}} \exp [-(B_1/\lambda^2) \sin^2 \theta] + F_{\mathrm{C,N,O,}} \exp [-(B_2/\lambda^2) \sin^2 \theta]\}$$

where n is a scale factor, F_{Br} and $F_{\mathrm{C,N,O,}}$ the bromine and light-atom contributions respectively calculated for atoms at rest, F the total structure factor, and B_1 and B_2 the two temperature factors. A least-squares procedure with B_1, B_2, and n as variables was employed to determine the values which would give the best agreement between calculated and observed structure factors. The values of B_1 and B_2 obtained were $B_1 = 2{\cdot}1$, $B_2 = 5{\cdot}0$. Fig. 1 shows the electron-density map for the [100] projection after the third stage of Fourier refinement. Fig. 2 shows a much less accurate electron-density map, based only on the bromine phases, of the [010] projection. The projection distance of 18·76 Å is so great that Fourier refinement on this projection is impossible because of the overlapping of atoms.

The proposed structure is therefore based mainly on the one refined projection. The second projection served to give the x co-ordinate of the bromine atom and to indicate the relative positions of the molecular components. The x co-ordinates of most of the atoms were derived by consideration of the molecular packing. The values of bond lengths and angles and van der Waals radii normally encountered in similar compounds were assumed to be valid in this case also. The various distances were adjusted and the x co-ordinates measured on an adjustable "wire spoke" model of the structure. The dimensions of the succinimide molecule have been determined by Mason [7] and were available in the later stages of this work. Independent measurements of the tetraethylammonium ion were obtained by the study of tetraethylammonium iodide (see following paper). The conformation [Fig. 1(*A*), following paper] of the ion is different in the two compounds but it is likely that the distortions of the $N{-}C_{(1)}{-}C_{(2)}$ bond angle observed in the iodide also occur in the molecular compound. A significantly better fit with the [100] projection was obtained by the use of a distorted model for the tetraethylammonium ion.

When the three-dimensional model had been constructed, some twenty intermolecular approach distances which had not been used as criteria for determining x co-ordinates were examined and were found all to be consistent with accepted values of van der Waals radii. This constitutes the main evidence for the correctness of the three-dimensional structure suggested.

Table 1 lists the estimated co-ordinates of the atoms as fractions of the unit cell edge. The y and z co-ordinates are derived from the refined [100] projection and are more accurate than the x co-ordinates which, with the exception of those of the bromide ion and $N_{(2)}$ are derived from packing studies.

Derived inter-atomic distances are not listed since many of these were assumed for the determination of the x co-ordinates. Table 2 lists calculated and observed structure factors for the [100] projection. The discrepancy factor $R = \sum||F_o| - |F_c||/\sum|F_o| = 0{\cdot}26$

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[6] Wilson, *Nature*, 1942, **150**, 152.

[7] Mason, *Acta Cryst.*, 1956, **9**, 405.

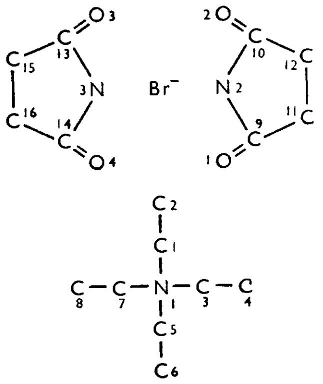

TABLE 1. *Atomic co-ordinates.*

	x	y	z		x	y	z
Br	0·175	0·025	0·134	$C_{(11)}$	$\overline{0{\cdot}100}$	0·068	0·227
$C_{(1)}$	0·067	0·2783	0·105	$C_{(12)}$	$\overline{0{\cdot}400}$	0·487	0·230
$C_{(2)}$	$\overline{0{\cdot}053}$	0·207	0·093	$N_{(2)}$	$\overline{0{\cdot}367}$	0·033	0·182
$C_{(3)}$	0·375	0·222	0·087	$O_{(1)}$	$\overline{0{\cdot}333}$	0·147	0·183
$C_{(4)}$	0·472	0·247	0·038	$O_{(2)}$	$\overline{0{\cdot}167}$	0·412	0·187
$C_{(5)}$	0·200	0·212	0·170	$C_{(13)}$	$\overline{0{\cdot}267}$	0·050	0·015
$C_{(6)}$	0·100	0·250	0·208	$C_{(14)}$	$\overline{0{\cdot}200}$	0·438	0·022
$C_{(7)}$	0·365	0·330	0·135	$C_{(15)}$	$\overline{0{\cdot}350}$	0·045	0·058
$C_{(8)}$	$\overline{0{\cdot}450}$	0·318	0·155	$C_{(16)}$	$\overline{0{\cdot}133}$	0·460	0·067
$N_{(1)}$	0·250	0·263	0·123	$N_{(3)}$	0·242	0·008	0·012
$C_{(9)}$	$\overline{0{\cdot}267}$	0·088	0·200	$O_{(3)}$	0·300	0·413	0·012
$C_{(10)}$	$\overline{0{\cdot}233}$	0·473	0·202	$O_{(4)}$	$\overline{0{\cdot}267}$	0·388	0·013

TABLE 2. *Observed structure amplitudes and calculated structure factors for hkl.* (In each set of three columns the first lists values of *l*, the second $F_{calc.}/10$, and the third $F_{obs.}/10$.)

00*l*

2	3·5	4·5	12	$\overline{6{\cdot}5}$	8·5	22	12	13·5	32	$\overline{2}$	2
4	$\overline{10{\cdot}5}$	15·5	14	14	12	24	$\overline{0{\cdot}5}$	3·5	34	5	5
6	29·5	28	16	19·5	16	26	$\overline{7{\cdot}5}$	10			
8	16	13	18	$\overline{9}$	11·5	28	$\overline{1{\cdot}5}$	3·5			
10	11	9·5	20	$\overline{5{\cdot}5}$	8	30	6·5	7			

02*l*

0	36·5	29·5	9	3	6·5	18	$\overline{9}$	9	27	1	1·5
1	$\overline{9{\cdot}5}$	9	10	$\overline{4{\cdot}5}$	8·5	19	1	2·5	28	$\overline{1}$	1·5
2	$\overline{2{\cdot}5}$	1	11	1·5	6	20	$\overline{5}$	6	29	1·5	1·5
3	0	3	12	$\overline{8}$	7	21	5	6	30	6·5	6
4	15·5	11	13	2	5·5	22	9·5	9	31	$\overline{1{\cdot}5}$	1·5
5	1·5	3·5	14	17·5	17	23	$\overline{1{\cdot}5}$	3	32	$\overline{1{\cdot}5}$	3
6	20·5	18·5	15	1	6	24	$\overline{2}$	6	33	0·5	0
7	$\overline{0{\cdot}5}$	4·5	16	18	17·5	25	$\overline{2}$	1·5	34	$\overline{5}$	5
8	19·5	14·5	17	$\overline{4{\cdot}5}$	6	26	$\overline{9{\cdot}5}$	6	35	1·5	0

04*l*

0	49·5	44	9	$\overline{22}$	17·5	18	$\overline{8}$	6	27	3	3·5
1	$\overline{13}$	15	10	$\overline{8}$	9·5	19	1	3	28	1	2·5
2	6·5	1	11	$\overline{5}$	0	20	$\overline{5}$	5	29	3	3
3	$\overline{10}$	5·5	12	$\overline{12{\cdot}5}$	13·5	21	4	6·5	30	6	5·5
4	$\overline{18}$	21	13	13·5	21	22	5	7·5	31	$\overline{3{\cdot}5}$	4·5
5	14·5	21	14	10	8	23	$\overline{5}$	3·5	32	$\overline{10}$	2·5
6	$\overline{5{\cdot}5}$	10·5	15	$\overline{2{\cdot}5}$	0	24	2·5	3	33	$\overline{2}$	2·5
7	9	12	16	4·6	3·5	25	$\overline{3{\cdot}5}$	3·5	34	$\overline{3{\cdot}5}$	2·5
8	14·5	10·5	17	8	5	26	$\overline{6{\cdot}5}$	8	35	2·5	0

06*l*

0	1·5	0	9	$\overline{14}$	17	18	$\overline{6{\cdot}5}$	5	27	2·5	3
1	$\overline{12{\cdot}5}$	14·5	10	$\overline{8{\cdot}5}$	12·5	19	$\overline{1{\cdot}5}$	0	28	0·5	0
2	$\overline{5{\cdot}5}$	6·5	11	$\overline{1{\cdot}5}$	1·5	20	$\overline{4}$	3·5	29	2·5	2
3	$\overline{13{\cdot}5}$	18	12	$\overline{6}$	3·5	21	8	9	30	3·5	3·5
4	$\overline{18{\cdot}5}$	18	13	5	4·5	22	5·5	5·5	31	$\overline{4{\cdot}5}$	4
5	$\overline{8}$	13	14	8·5	8	23	$\overline{5}$	5·5	32	$\overline{0{\cdot}5}$	0
6	0	4·5	15	$\overline{6{\cdot}5}$	4·5	24	0	0	33	$\overline{2}$	0
7	$\overline{5}$	0	16	4·5	4·5	25	$\overline{7{\cdot}5}$	9	34	$\overline{2{\cdot}5}$	2·5
8	8	9	17	$\overline{14{\cdot}5}$	8·5	26	$\overline{6}$	4·5			

08*l*

0	2·5	7·5	9	$\overline{16}$	17·5	18	$\overline{2{\cdot}5}$	1·5	27	$\overline{4{\cdot}5}$	6
1	$\overline{15{\cdot}5}$	11	10	$\overline{2{\cdot}5}$	0	19	$\overline{0{\cdot}5}$	1·5	28	1	0
2	$\overline{2{\cdot}5}$	1	11	$\overline{12}$	8·5	20	$\overline{1}$	0	29	2	1·5
3	$\overline{11{\cdot}5}$	10·5	12	$\overline{3{\cdot}5}$	5·5	21	6·5	7	30	2	0
4	$\overline{6{\cdot}5}$	1·5	13	6·5	8·5	22	1·5	1·5	31	$\overline{4}$	2·5
5	16	17·5	14	$\overline{1}$	0	23	$\overline{8{\cdot}5}$	8	32	3	0
6	$\overline{3{\cdot}5}$	6·5	15	1·5	2·5	24	1·5	1·5	33	$\overline{3{\cdot}5}$	1·5
7	$\overline{3}$	3·5	16	3	2	25	$\overline{5{\cdot}5}$	6·5			
8	0	6·5	17	16	13·5	26	$\overline{2}$	2·5			

TABLE 2. (*Continued.*)

010*l*

0	$\overline{4}$	3·5	8	$\overline{1}$	2	16	$\overline{1{\cdot}5}$	3·5	24	0·5	2·5
1	$\overline{13{\cdot}5}$	13	9	$\overline{13{\cdot}5}$	13·5	17	$\overline{14{\cdot}5}$	11	25	$\overline{7}$	8
2	$\overline{1{\cdot}5}$	1·5	10	2	6	18	$\overline{1}$	2·5	26	0	0
3	$\overline{14{\cdot}5}$	14	11	$\overline{1{\cdot}5}$	3·5	19	1·5	0	27	4·5	6
4	$\overline{4{\cdot}5}$	6·5	12	3·5	5·5	20	0·5	0	28	$\overline{0{\cdot}5}$	3·5
5	$\overline{17{\cdot}5}$	19	13	8·5	6	21	9·5	11	29	2·5	0
6	$\overline{3}$	0	14	1·5	0	22	0·5	0	30	0	3
7	$\overline{2}$	2·5	15	$\overline{3}$	0	23	$\overline{5{\cdot}5}$	4·5	31	3·5	0

012*l*

0	$\overline{5}$	4·5	8	$\overline{4}$	2·5	16	$\overline{0{\cdot}5}$	3	24	$\overline{1}$	0
1	$\overline{5}$	4·5	9	$\overline{11{\cdot}5}$	11·5	17	$\overline{8{\cdot}5}$	7	25	$\overline{5}$	5·5
2	$\overline{4{\cdot}5}$	5·5	10	1·5	0	18	1·5	0	26	3	3·5
3	$\overline{10{\cdot}5}$	9	11	$\overline{5}$	3·5	19	2·5	1·5	27	4·5	4·5
4	3·5	6	12	$\overline{3}$	3·5	20	2	0	28	0	3·5
5	10·5	11	13	9	11	21	6·5	4·5	29	3·5	3
6	0·5	1·5	14	$\overline{4}$	4·5	22	$\overline{2{\cdot}5}$	3	30	$\overline{1}$	2·5
7	5·5	8	15	0	0	23	$\overline{3{\cdot}5}$	0			

014*l*

0	$\overline{10}$	10·5	8	$\overline{6}$	6·5	16	$\overline{3{\cdot}5}$	8	24	$\overline{0{\cdot}5}$	2
1	$\overline{5}$	4	9	$\overline{8{\cdot}5}$	6·5	17	$\overline{7}$	5·5	25	$\overline{3{\cdot}5}$	2·5
2	0	3	10	5·5	6	18	4·5	4·5	26	3·5	2·5
3	$\overline{5}$	4	11	$\overline{0{\cdot}5}$	0	19	2·5	2·5	27	3·5	3
4	5	6·5	12	7·5	8·5	20	1	0	28	$\overline{0{\cdot}5}$	0·5
5	10	6	13	8	3·5	21	6	3·5	29	2·5	0
6	$\overline{4}$	6	14	$\overline{5{\cdot}5}$	7	22	$\overline{3}$	2			
7	2	2·5	15	$\overline{0{\cdot}5}$	0	23	$\overline{2}$	2·5			

016*l*

0	$\overline{12{\cdot}5}$	11	7	2·5	3	14	$\overline{4{\cdot}5}$	1·5	21	4	0
1	$\overline{6}$	7	8	$\overline{3{\cdot}5}$	0	15	0	1·5	22	$\overline{5}$	4·5
2	$\overline{0{\cdot}5}$	4·5	9	$\overline{4{\cdot}5}$	5·5	16	$\overline{5{\cdot}5}$	5·5	23	1	0
3	$\overline{4{\cdot}5}$	4·5	10	1	0	17	$\overline{4}$	5	24	0·5	0
4	7·5	11	11	$\overline{1{\cdot}5}$	3·5	18	5	5·5	25	$\overline{2{\cdot}5}$	3·5
5	7	7	12	2·5	3	19	2	0	26	4·5	4
6	$\overline{3{\cdot}5}$	3·5	13	5	3	20	2	0	27	1·5	0

018*l*

0	$\overline{11}$	5·5	7	1·5	0	14	$\overline{4}$	2·5	21	$\overline{1{\cdot}5}$	0
1	0·5	0	8	$\overline{5}$	0	15	0	0	22	4	3
2	1·5	4·5	9	$\overline{2{\cdot}5}$	1·5	16	$\overline{5}$	5·5	23	$\overline{1}$	0
3	0·5	0	10	4	3·5	17	1	0	24	0·5	0
4	6	7	11	$\overline{0{\cdot}5}$	0	18	5·5	5·5	25	$\overline{1}$	0
5	2	0	12	6·5	6·5	19	0·5	0			
6	$\overline{3{\cdot}5}$	1·5	13	2·5	0	20	2·5	2			

020*l*

0	$\overline{8}$	6	6	1·5	0	12	5	3	18	4·5	2·5
1	$\overline{1}$	0	7	0·5	0	13	0·5	0	19	$\overline{0{\cdot}5}$	0
2	1·5	3·5	8	$\overline{5{\cdot}5}$	3·5	14	$\overline{4}$	3	20	2	1
3	$\overline{0{\cdot}5}$	0	9	0	0	15	$\overline{0{\cdot}5}$	0			
4	7·5	8·5	10	4	3·5	16	$\overline{3}$	2			
5	1	0	11	0·5	0	17	0·5	0			

022*l*

0	$\overline{6}$	3·5	4	4·5	3	8	$\overline{4{\cdot}5}$	3·5	12	4	2·5
1	2	2·5	5	$\overline{1}$	1·5	9	1·5	2	13	$\overline{1{\cdot}5}$	0
2	0·5	0	6	$\overline{1}$	0	10	3	1·5	14	$\overline{3}$	2
3	2	0	7	$\overline{0{\cdot}5}$	2	11	0	0	15	0·5	0

024*l*

0	$\overline{4}$	1	2	0·5	0	4	3·5	1	6	$\overline{1{\cdot}5}$	0·5
	12	1·5	3	1·5	0	5	$\overline{2{\cdot}5}$	1·5			

which is not unsatisfactory for a structure of this complexity. Fig. 3 gives a general view of the structure down [100].

The groups of atoms identified in the structure and their dispositions relative to each other are such that the molecular formula proposed by Braude and Waight [5] must be rejected. The tetraethylammonium and bromide ions and the succinimide molecule retain their individuality in the structure. All like ions are structurally equivalent but the succinimide molecules are of two crystallographically different kinds. The structure as a whole may be considered in relation to the cohesion between its three components.

The shortest distances between any of them are those (3·4 Å) between the bromide ions and the nitrogen atoms of succinimide molecules. Compared with 3·3 Å for the bromide ion to OH distance in sucrose–sodium bromide, this suggests a weak hydrogen bond. Otherwise the components are at distances corresponding to ionic or van der Waals radii. The only distinguishable complex of finite size formed by the three components is this group of one bromide ion and two succinimide molecules, one of each crystallographic kind. If this is regarded as a form of complex negative ion the structure as a whole consists of sheets of tetraethylammonium ions parallel to (010) carrying a positive charge and spaced at $b/2$ or about 9·9 Å from each other. The negatively charged complexes form parallel sheets between them.

Alternatively, a description may begin with the electrostatic interaction of bromide and tetraethylammonium ions. The latter have roughly the form of oblate spheroids.

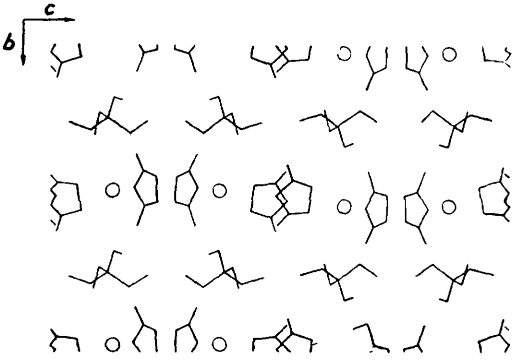

FIG. 3. *The structure as seen along the a axis. The origin is at the centre of the figure.*

(Circles = bromine ions. Branched pentagons, some overlapping in pairs, = succinimide molecules. Insect-like figures show the C–C and C–N bonds of tetraethylammonium ions.)

They are arranged with their short axes roughly parallel to b. Each has two bromide ions in the general directions of its minor axes, one on each side at a distance of about 4·7 Å. Sheets of tetraethylammonium are joined by negative bromide ions which lie roughly above and below the positive centres. The bromide ions have diameters considerably smaller than the major axis of the tetraethylammonium spheroid and therefore cannot completely fill the sheet of the same area. In this sense the succinimide molecules may be said to fill the spaces left by the arrangement of the positive and negative ions.

Another description emphasises the interaction of tetraethylammonium and succinimide. The oxygen atoms of the succinimide molecules will carry some negative charge, and both oxygen atoms of both kinds of succinimide molecule are found to be directed towards a positive tetraethylammonium ion. In this way succinimide molecules form bridges linking the different positive sheets. There is a certain coherence of the complex ions and succinimide molecules to form a three-dimensional extended structure which as a whole is positively charged. The negative ions may be described as placed in the holes of this structure. From this point of view the behaviour in the series of halides is explicable. The bromide ion just fits the hole. The smaller chloride ion therefore does not completely fill the space and the deliquescence of the compound containing it may be connected with the consequent instability. The iodide ion is too large for the hole and the corresponding compounds could not be prepared.

The general constructional principle of the whole is therefore that of local neutralisation of charges; the positive NH part of succinimide is directed towards the negative ions, the negative oxygens of the succinimide towards the positive ions, and the negative and positive ions themselves are in close contact.

The structure is related to " co-ordination structures " in V. M. Goldschmidt's sense.

Complete co-ordination of the various components, with the limitations imposed by their dimensions, is apparently achieved only by having two crystallographically different sets of succinimide molecules. Thus all four CO groups directed towards the tetraethylammonium ion are geometrically different although the nature of their attachment is the same. That a particular pair of substances, tetraethylammonium bromide and succinimide, should form an addition compound at all is seen to be dependent both on the distribution of charge in their components and on accidental and scarcely predictable dimensional factors.

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