X-Ray Studies on the Oxyhalide Minerals Nadorite (Ochrolite) *PbSbO₂Cl* and Ekdemite.

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Recent investigations have revealed a family of very simply-built oxyhalides, containing bismuth together with another metal. These compounds all form tetragonal plates; metal-oxygen sheets, which have in all substances the same construction, extend parallel to the largest faces. The Me-O-sheets are separated by halogen sheets, which may be single (X_1-compounds), double (X_2) or triple (X_3). The tetragonal a-axis is always close to 3.9 Å, which is easily explained from the structure of the Me-O-layers (Sillén, 1940).

We thought it of interest to search for the same simple construction principles in oxyhalides containing no bismuth. As first objects of study we chose the minerals nadorite, $PbSbO_2Cl$, and ekdemite, a lead-arsenic oxychloride for which several different formulae have been proposed. The crystal structure of nadorite was worked out, and proved to be, in principle, an X_1 structure, where however the two sorts of metal atoms are ordered in rows, whereby the symmetry descends to orthorhombic. It could be shown that the mineral named ochrolite is identical with nadorite.

Our work on ekdemite is also recorded. Here the conditions are more complex, and no certain conclusions could be drawn.

Syntheses in the system *PbSbOCl*. Tetragonal *PbSbO₂Cl*.

Lead chloride was fused together with Sb_2O_3, mixtures of PbO and Sb_2O_3 in various proportions, or PbO. Series of samples were prepared in porcelain crucibles and in evacuated tubes of Supremax glass; in both cases one series with rapid and one with slow cooling of the melts was made. The cooled product was ground and, for removal of the surplus of $PbCl_2$, brought into a large beaker with cool water. After 5—10 hours of electric stirring the insoluble residue was filtered off and dried in a cool vacuum desiccator. The powder photographs revealed in these samples 1) a tetragonal phase, 2) nadorite, which is orthorhombic, 3) a phase which was obtained pure when PbO was added to $PbCl_2$ and which consequently contains no antimony. It seems improbable that the two first phases should contain antimony of higher valency than three, as they were obtained also

in evacuated tubes. In the powder photographs of some samples a few weak lines appeared together with the lines of the tetragonal phase, indicating that there may be still more phases, presumably richer in antimony.

According to the powder photographs of the tetragonal phase, it is isomorphous with the X_1 compounds[1]), of formula Me_2O_2X (Sillén, 1939), and thus its formula should be $PbSbO_2Cl$, or the same as that of nadorite. It seems very improbable that there should be empty points in the lattice or that Cl^- or O^{2-} should be partially replaced by OH^-, as the cell dimensions in six samples were found to be the same within the limits of accuracy.

Thus there seem to be two compounds of formula $PbSbO_2Cl$, one tetragonal and one orthorhombic. Their stability conditions were not studied in detail. Nadorite appeared at the calculated composition, $(Sb_2O_3 + PbO)$ + surplus of $PbCl_2$, and in samples richer in PbO. The tetragonal phase was formed whenever Sb_2O_3 was in excess over PbO in the melt. If synthetic nadorite was boiled with water, the tetragonal phase was formed.

The cell dimensions of tetragonal $PbSbO_2Cl$ were found to be $a = 3.887$Å, $c = 12.26$ Å. As the c-axis happens to be close to $a\sqrt{10}$, different powder lines frequently overlap. Therefore the metal parameter could not be estimated from the intensities more accurately than to $z = .16 \pm .01$. The two most probable regions are $.153 \pm .003$ and $.168 \pm .003$, between which no certain decision could be made.

Nadorite.   Unit cell.

Samples of natural nadorite were available from the Långban mine in Värmland, Sweden, and from Djebel Nador, Constantine, Algeria. The Nador sample consisted of brown transparent crystal plates with a dinstinct rifling; in the Långban sample, which was yellow, there were no so well-shaped single crystals. The powder photographs of these two specimens were identical, as far as could be seen, but differed very slightly from those of the synthetic nadorite (see below). Algerian nadorite has been analyzed by Flajolot (1870), Pisani (1870), and Tobler (1872), who all agree upon the formula $PbSbO_2Cl$.

Two crystal plates were picked out from the Nador sample and set. The Laue symmetry proved to be D_{2h}-mmm. It was decided to denote the axis perpendicular to the crystal plates by c, and the one parallel with the rifles by b. This choice of axes will later on prove suitable for the comparison with the tetragonal X_1 compounds though it does not agree with the description in the "International Tables".

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1) Atomic positions: D_{4h}^{17} — $I/4\,mmm$ $(0\ 0\ 0, \frac{1}{2}\frac{1}{2}\frac{1}{2})$ +, $4\,Me$ in $4\,(e)$: $\pm (0\ 0\ z)$, $4\,O$ in $4\,(d)$: $0\frac{1}{2}\frac{1}{4}$, $\frac{1}{2}0\frac{1}{4}$, $2\,X$ in $2\,(b)$: $0\ 0\frac{1}{2}$.

Rotation photographs around the a and b axes, and Weissenberg photographs of $h0l$, $h1l$, $0kl$, and $1kl$ were taken. The cell dimensions

$$a = 5.59_1 \text{ Å} \qquad b = 5.43_1 \text{ Å} \qquad c = 12.20 \text{ Å}$$

explain the powder photographs of the two natural samples (Tab. I) very well. For the synthetic nadorite a slightly different set of axes:

$$a = 5.58_5 \text{ Å} \qquad b = 5.43_1 \text{ Å} \qquad c = 12.21 \text{ Å}$$

Table I. Powder photographs of natural nadorite, $PbSbO_2Cl$. $CrK\alpha$ radiation.

$h\,k\,l$	$\sin^2\theta$ calc.	$\sin^2\theta$ obs.	I obs.	$h\,k\,l$	$\sin^2\theta$ calc.	$\sin^2\theta$ obs.	I obs.
012	.0794	.0791	(w)	026	.4933	.4935	w
111	.0949	.0947	m	313	.4995	.5000	m
103	.1208	.1203	w	117	.5163	.5165	m
004	.1405	.1402	w	133	.5195	.5205	m
113	.1651	.1655	st				
200	.1672	.1683	w	216	.5276	.5279	m
020	.1772	.1777	w	034	.5392	(.5393	vw)
014	.1848	.1848	vvw	008	.5619⎫	.5616	vw
121	.2278	.2286	vvw	321	.5622⎭		
				232	.6010	(.6022	vvw)
212	.2466	(.2469	w)	323	.6324	.6330	vw
105	.2613	(.2625	w)	315	.6400	.6410	vvw
123	.2980	(.2981	m)	127	.6492	.6495	w +
115	.3056⎫	.3074	m	135	.6600⎫	.6607	w +
204	.3077⎭			226	.6605⎭		
006	.3161⎫	.3172	m	400	.6688	.6697	w +
024	.3177⎭			234	.7064	.7054	vvw
220	.3444	.3446	m	040	.7088	.7086	w
214	.3520	.3518	vw	036	.7148	.7145	vw
016	.3604	.3599	m	208	.7291	.7293	vw
222	.3795	.3808	vvw	028	.7391	.7398	vvw
301	.3850	.3857	vvw	412	.7482	.7487	vvw
311	.4293	(.4300	w)	109	.7530	.7522	w
125	.4385	(.4380	vw)	331	.7837	.7841	vw
131	.4492	.4495	w	307	.8064	.8057	vw
303	.4552	.4560	vw	404	.8093	.8089	w
107	.4720	.4720	w	143	.8296	.8295	vw
206	.4833⎫	.4845	m	420	.8460	.8460	vvw
224	.4849⎭			044	.8492	.8490	w

gives a better agreement. The deviations, the directions but not the amounts of which are certain, may depend upon small contaminations, e. g. of arsenic, in the minerals.

The ratio of the axes in the mineral $a:b:c = 1:.971:2.181$ agrees well with the measurements of Césaro (1888), suitably rearranged: $c:b:3a$ $= 1:.9699:2.1792$. If the cell contains 4 $PbSbO_2Cl$, the density should be 7.05, which agrees well with the density 7.02, measured by Pisani (1870).

It is apparent that the a and b axes of nadorite are close to 5.49 Å, the a axis of tetragonal $PbSbO_2Cl$ multiplied with $\sqrt{2}$, and that the c axes of the two phases are almost identical. Accordingly the cell volume of nadorite, 370.5 Å³, is almost exactly twice that of the tetragonal compound, 185,3 Å³; the former contains 4 $PbSbO_2Cl$, the latter 2 $PbSbO_2Cl$. One might thus expect, as was later proved, that the crystal structure of nadorite is mainly the same as that of tetragonal $PbSbO_2Cl$, and especially that the bulky anions will be very similarly arranged in the two structures.

Nadorite. Atomic arrangement.

The following classes of reflections were absent: $hk0$ with k odd and hkl with $(h + l)$ odd (accordingly $h0l$ with $(h + l)$ odd, $0kl$ with l odd, $hk0$ with h odd). These extinctions are characteristic of the space-groups D_{2h}^{17}, C_{2v}^{12} and C_{2v}^{16}. The holohedral D_{2h}^{17} was first tried; as a plausible crystal structure could be devised within this space-group there was no need to descend to a lower symmetry. The axes a, b, and c in our description correspond to a, c, and b in the description of D_{2h}^{17} in the "International Tables".

For the 4 Pb and 4 Sb in a unit cell there are in D_{2h}^{17} three fourfold point positions: 4 (a) und 4 (b), which have no parameter, and 4 (c): $0 \frac{1}{4} z$, $0 \frac{3}{4} \bar{z}$. The observed intensities in the Weissenberg photographs made it highly improbable that any metal atoms should occupy parameter-free points. It was preliminarily assumed that the metal atoms occupy two positions 4 (c) with the parameters z_1 for Pb and z_2 for Sb.

The distances between metal atoms would then cause maxima in the Patterson function at the following points: $0 \frac{1}{2} 2z_1$; $0\,0\,(z_1 - z_2)$; $0 \frac{1}{2} (z_1 + z_2)$; and $0 \frac{1}{2} 2 z_2$. The relative weights of these maxima would be $Pb^2 : 2\,PbSb : 2\,PbSb : Sb^2$ or about $8:10:10:3$. One can expect to discern all except the last in suitable cuts of the P function (see Sillén 1940, 1941), namely

$$P\,(p0z) \approx \sum_l F^2\,(00l)\cos 2\pi l z + \text{const.} \sum_l F^2\,(01l)\cos 2\pi l z$$

$$P\,(p\tfrac{1}{2}z) \approx \sum_l F^2\,(00l)\cos 2\pi l z - \text{const.} \sum_l F^2\,(01l)\cos 2\pi l z$$

$$P\,(0pz) \approx \sum_l F^2\,(00l)\cos 2\pi l z + \text{const.} \sum_l F^2\,(10l)\cos 2\pi l z.$$

These formulae are valid because all heavy atoms are in 4 (c) positions; the constants should have unit order of magnitude.

The sums were calculated — inserting however the estimated intensities instead of the F^2 — and plotted against z for $0 \leq z \leq .50$. By comparison of the curves maxima were found in $P (0pz)$ at .20 and .30, and in $P (p \frac{1}{2} z)$ at .05, .25, and .45. The ambiguity of these cuts, which are symmetrical around .25, was removed by the maxima in $P (0pz)$ at .25, .30 and .45. By comparison with the expected positions of maxima we could conclude that $z_1 - z_2 \approx .30$ and that one of $z_1 + z_2$ and $2 z_1$ is .25 or .75 and the other .45 or .55. It is always possible to choose such an origin of coordinates that $z_1 < z_2$ and $z_2 < .25$. Then the Patterson maxima found can be explained only by the values $z_1 \approx .375$ and $z_2 \approx .075$.

With these parameters, the metal ions in nadorite will form double layers very similar to those in the tetragonal X_1 compounds, although in nadorite there are two classes of metal ions: the Sb somewhat closer to and the Pb more distant from the central plane of each layer. It seemed natural to assume that just as in the tetragonal X_1 compounds each double metal layer contains a sheet of oxygen atoms and that the metal-oxygen layers are separated by single sheets of halogen ions. A different arrangement of the anions would, moreover, hardly be possible, for spatial reasons.

As a first approximation, it was assumed that the anions occupy "ideal" positions: $4Cl$ in 4 (c), $z_x = \frac{3}{4}$ and $8 O$ in 8 (e), $x = \frac{1}{4}$. The metal parameters were varied, and the best agreement with observed intensities was found with $z_1 = .380 \pm .002$, $z_2 = .078 \pm .005$. The positions of the anions could not be determined accurately from the intensity data.

It was considered that the chlorine parameter is probably $z_x = .756 \pm .010$, as with this value the distances from a chlorine ion to the four surrounding lead ions become equal. We could draw no certain conclusions about the deviation of the oxygen parameter from .25. With the ideal oxygen parameter, the $Sb - O$ distance in nadorite would be 2.17 Å (coordination $Sb - 4 O$), whereas according to Buerger and Hendricks (1937) $Sb - O = 2.00$ Å ($Sb - 3 O$) in valentinite. (Bozorth who in 1923 calculated $Sb - O = 2.22$ Å in senarmontite has afterwards stated that this value is not very accurate.) If the oxygen parameter x in nadorite is decreased, the distance $Sb - O$ will certainly decrease, but at the same time the distances $O - O$ along the a axis will alternately be above and below the rather normal value $a/2 = 2.80$ Å. A larger deviation from .25 than .02 — making the said $O - O$ distances 2.57 and 3.02 Å — was thought improbable.

The change in the chlorine parameter affected the structure factors so little that there was no reason to shift the metal parameters. In Tab. II the final values of 40 $(F/4 f_{Pb})^2$ are compared with the observed intensities of 00l, 01l, 10l, 11l and 20l (02l). The observed intensities are averaged from different photographs; e. g. the spots 10l appear in the $h0l$ and $1kl$ Weissenberg photographs with different intensity because of the difference in shape and position of the crystals. The agreement is as good as can be expected in view of the general uncertainty in the atomic scattering factors. I_{calc} (018), which is zero, can be made reasonable by small displacements within the uncertainty region. The intensities of different lines in the powder photographs cannot be compared directly because of a certain orientation effect.

Table II. Calculated and observed intensities in Weissenberg photographs of nadorite. $Cu\,K$ radiation.

| 00l | | 01l | | 02l (20l) | | 10l | | | 11l | |
l	calc. obs.	calc.	obs.	calc.	obs.	l	calc.	obs.	calc.	obs.
0		0	—	97	m	1	2	vvw	22	w
2	6 w	12	vw	0	—	3	19	w	95	m
4	30 m	8	vw	58	m	5	6	vw	6	vvw
6	24 m	45	m	45	m	7	52	m	29	m
8	28 m	0	vw	14	w	9	40	m	2	vw
10	5 w	100	st	1	vw	11	22	w	14	w
12	1 —	11	w	5	vw	13	101	st	8	vw
14	0 —	52	st	1	vw	15	1	—	4	vw
16	48 (βvw)	53	(βvw)	35	(βvw)	17	52	(βw)		

The complete crystal structure of nadorite would thus be:
D_{2h}^{17} — $Bmmb$ (000, $\frac{1}{2}$ 0 $\frac{1}{2}$) +

$$
\begin{array}{lll}
4\ Pb \text{ in } 4\,(c) & 0\ \frac{1}{4}\ z,\ 0\ \frac{3}{4}\ \bar{z} & z_1 = .380 \pm .002 \\
4\ Sb \text{ in } 4\,(c) & & z_2 = .078 \pm .005 \\
4\ Cl \text{ in } 4\,(c) & & z_x = .756 \pm .010 \\
8\ O \text{ in } 8\,(e) & \pm(x\,0\,0,\ x\,0\,\frac{1}{2}) & x = .25 \pm .02
\end{array}
$$

In Fig. 1 two unit cells of this structure are depicted. The interatomic coordination and distances in nadorite will be: $Pb - 4\,O$ $(O - 4\ Pb)$ 2.44 Å, $Pb - 4\ Cl$ $(Cl - 4\ Pb)$ 3.18 Å, $Sb - 4\,O$ $(O - 2\ Sb)$ 2.17 Å, $Sb - 4\ Cl$ $(Cl - 4\ Sb)$ 3.39 or 3.54 Å, $O - 4\,O$ 2.80 Å (a direction) or 2.72 Å (b direction), $O - 4\ Cl$ $(Cl - 8\,O)$ 3.56 and 3.68 Å, and $Cl - 4\ Cl$ 3.90 Å. In tetragonal $PbSbO_2Cl$ we have $Pb(Sb)\ -4\,O$ $(O - 4\ Me)$ 2.2_3 Å, $Pb(Sb) - 4\ Cl$ $(Cl - 8\ Me)$ 3.3_8 Å, $O - 4\,O$ 2.75 Å, $O - 4\ Cl$ $(Cl - 8\,O)$ 3.63 Å and $Cl - 4\ Cl$ 3.89 Å.

The arrangement of anions and cations is in principle the same in nadorite and tetragonal $PbSbO_2Cl$. In nadorite, the Pb and Sb atoms are
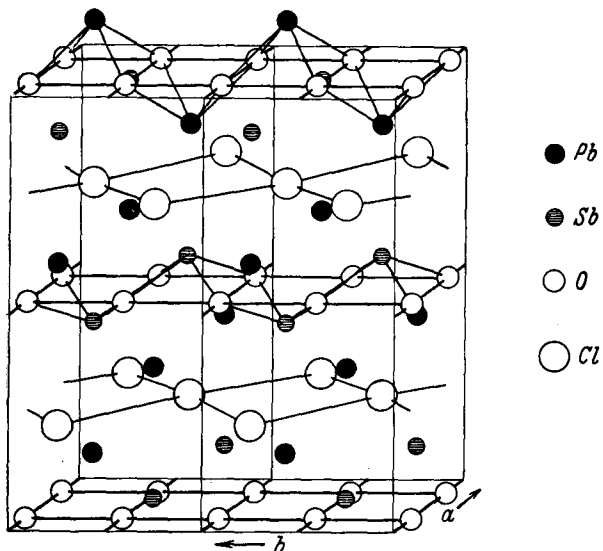

Fig. 1. Two adjacent unit cells of nadorite, $PbSbO_2Cl$. The chlorine and oxygen sheets, as well as certain lines of contact between oxygen and metal atoms, have been marked out.
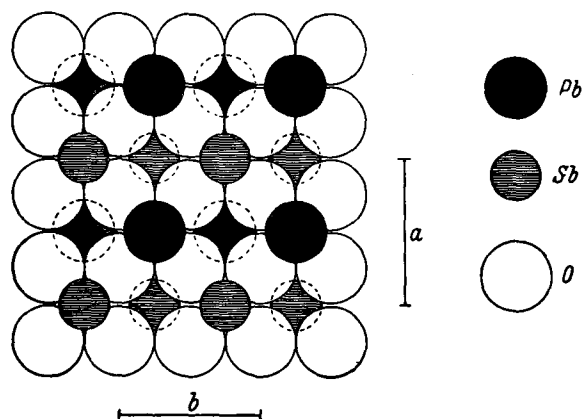

Fig. 2. A metal-oxygen sheet in nadorite, viewed along the c-axis. In the tetragonal X_1 compound, the Pb and Sb atoms are distributed at random over the metal positions.

not randomly mixed in the same point position but are separated in different rows (Fig. 2). The lead atoms have moved in direction from the oxygen to

the halogen sheets, making both distances $Pb - O$ and $Pb - Cl$ more ordinary. The antimony atoms in nadorite have approached the oxygen sheets, apparently losing all contact with the chlorine atoms.

It is easily understood that nadorite will have its best cleavage perpendicular to the c-axis, that is in the plane of the oxygen and halogen sheets. It may be added that, according to the Weissenberg photographs, there was in each of the two single crystals a very small part where the a and b axes were at right angles to those in the major part of the crystal. Thus exceedingly weak spots were found at the places of the (missing) 1.0.10, 1.0.14 and 0.1.13, evidently corresponding to the strong reflections 0.1.10, 0.1.14 and 1.0.13 from the parts with different crystal orientation.

Ochrolite.

Flink (1889) described a yellow orthorhombic mineral which he named ochrolite and to which he attributed the formula $Pb_6Sb_2O_7Cl_4$. As, however, our syntheses revealed no lead-antimony oxychloride richer in lead than nadorite, we soon suspected that ochrolite is identical with nadorite.

Flink once worked at the Mineralogical Department of the Stockholm University. By the courtesy of Professor P. Quensel we obtained from its collections the single crystal of "ochrolite", on which Flink had carried out his goniometric measurements. By the oscillation method of Arnfelt (1939) we set it for rotation around one of the two shorter crystal axes. Weissenberg photographs were taken of the equator zone and of the first layer line; they proved to be exactly identical with the $h0l$ and $h1l$ photographs of nadorite.

It can thus safely be said that the mineral ochrolite is identical with nadorite, the existence of which was probably not known to Flink. As Flink had barely a hundred mg of the mineral, moreover impure, an error in his analysis is easily understood. The formula proposed by him was apparently influenced by analogy with ekdemite.

Ekdemite.

Nordenskiöld (1877) discovered at Långban a new mineral, for which he proposed the name ekdemite. According to Nordenskiöld, it crystallizes in uniaxial, presumably tetragonal, yellow plates and has a composition close to $Pb_7As_2O_8Cl_4$. He observed also an orthorhombic mineral, containig the same elements as ekdemite, but could not get a suffi-

cient amount of it for an analysis. Flink (1888) found at Pajsberg a biaxial, orthorhombic mineral, of formula $Pb_6As_2O_7Cl_4$, which he called heliophyllite.

Hamberg (1889) investigated a series of samples labelled as ekdemite or heliophyllite from Harstigen, close to Pajsberg. In every sample he found uniaxial as well as biaxial lamellae, intimately grown together. The samples could be divided into two classes; "type 1" containing about equal amounts of uni- and biaxial lamellae, and "type 2" consisting mainly of the uniaxial mineral, with streaks of the biaxial one. Analyses of samples of "type 1" and "type 2" differed very little. Hamberg thus proposed a common formula, $Pb_{13}As_4O_{15}Cl_8$ for the uniaxial substance (ekdemite) and for the biaxial one. He thought it probable that the latter had been formed by external pressure upon uniaxial ekdemite, and should therefore be considered as a variety of ekdemite; if it were to be an independent mineral, it should be named heliophyllite.

We had obtained two specimens of ekdemite from Harstigen and one from Jakobsberg. (All these places — Långban, Pajsberg, Harstigen and Jakobsberg — are situated in the Swedish province of Värmland.) All three samples were yellow and had a distinct plane of cleavage. Out of one of the Harstigen samples a table was cut, on which Laue, rotation and Weissenberg photographs were taken. It revealed as far as could be seen a perfect tetragonal symmetry, $4/mmm$; the cell dimensions seemed to be $a = b = 10.8$ Å, $c = 25.6$ Å, which also agreed with the powder photographs. However plates out of this sample proved under the polarizing microscope to be biaxial. As in large tables different parts were found to have axial planes perpendicular to each others, a very small plate was set which was certainly homogeneous. In its Weissenberg photographs no difference was seen from those of the larger crystal plate, except that the spots were naturally rather weak. The deviations from tetragonal symmetry can thus appear only in the very weakest spots.

In the two other ekdemite samples, the powder photographs revealed the existence of still another phase besides the pseudo-tetragonal one. The optical investigation accordingly showed that the crystal plates contain uniaxial parts as well as biaxial ones, the latter with two mutually perpendicular orientations of the axes. In none of our samples was the uniaxial phase predominant; therefore no X-ray investigation of it was attempted.

According to Professor P. Quensel, the biaxial mineral is optically negative with a strong dispersion, $\varrho < v$, and a rather low birefringence. The uniaxial one is also optically negative and shows a considerably larger birefringence.

As for the chemical composition, we had expected $Pb_3AsO_4Cl_2$, with four-valent (or rather equal amounts of three- and five-valent) arsenic. This formula would agree with the general formula, Me_2O_2X, of the X_1 phases and would also agree well with the analyses of previous authors. However, the presence of arsenic of a higher valency than three could be excluded by a simple test. In a test tube a small amount of ekdemite (pure biaxial from Harstigen) was dissolved in hot concentrated HCl; the solution was diluted with the double volume of distilled water and boiled free from air. In another test tube an about equal amount of $(As_2O_3 + As_2O_5)$ was treated in the same way, and in a third tube a blind test with HCl and water was made. To each of the three tubes was added a boiled aqueous solution of KI. The As_2O_5 solution was coloured deep brown by liberated iodine; the ekdemite and blind test solutions remained practically colourless.

We tried to synthesize ekdemite by melting As_2O_3, PbO, or mixtures of them, with an excess of $PbCl_2$ in Supremax tubes as described for nadorite. In eight different samples three phases were found (except the arsenic-free one obtained with pure $PbO + PbCl_2$). In another six samples ($As_2O_3 + As_2O_5$), or As_2O_5 was used instead of As_2O_3. Still another phase appeared. For none of these four phases the powder photographs were even similar to those of the two different ekdemite phases.

The failure to synthesize ekdemite as well as the somewhat awkward formula arrived at make us suspect that something essential may have been overlooked at previous analyses. As, however, the crystal structures of both phases seem to be rather complicated, and as only small amounts of the mineral are available, we decided to leave the ekdemite problem to the skill of future investigators.

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