Summary. Yellow sapphirine with a doubled b-axis occurs in enstatite-hornblende-sapphirine rocks at the contact between dolomite marble and yoderite-quartz-talc schist on Mautia Hill, Tanganyika. It has refractive indices $\alpha 1.725 \pm 0.002$, $\gamma 1.732 \pm 0.002$, $2V 66^\circ$, pleochroism $\alpha$ pale yellow, $\beta$ pale lime green, $\gamma$ pale pinkish orange, and a composition corresponding to

$$\text{Mg}_{5.60}\text{Mn}_{0.04}\text{Fe}^{2+}_{0.17}\text{Ti}_{0.01}\text{Fe}^{3+}_{0.35}\text{Al}_{0.07}\text{Si}_{1.75}\text{O}_{20.08}.$$ 

Its unit-cell dimensions are approximately $a 9.85 \AA$, $b 28.6 \AA$, $c 9.96 \AA$, $\beta 110^\circ 30'$, and its diffraction pattern exhibits diffuse streaks parallel to $b^*$, on which lie diffuse intensity maxima representing the relics of certain types of sharp reflexions of normal sapphirine; this is interpreted as indicative of partial MgAl ordering. The associated enstatite and hornblende have compositions

$$\text{Mg}_{8.30}\text{Mn}_{0.02}\text{Fe}^{2+}_{0.05}\text{Ti}_{0.01}\text{Fe}^{3+}_{0.13}\text{Al}^{[6]}_{0.25}\text{Al}^{[4]}_{0.41}\text{Si}_{7.39}\text{O}_{24.00}$$

and

$$\text{K}_{0.04}\text{Na}_{0.10}\text{Ca}_{1.32}\text{Mg}_{4.37}\text{Mn}_{0.01}\text{Fe}^{2+}_{0.03}\text{Ti}_{0.04}\text{Fe}^{3+}_{0.04}\text{Al}^{[6]}_{0.30}\text{Al}^{[4]}_{0.10}\text{Si}_{7.39}\text{O}_{22.24} (\text{OH})_{1.76}$$

respectively and are accompanied by accessory hematite and pseudobrookite. Preliminary heating experiments have demonstrated the transformation of single crystals of partially ordered Mautia sapphirine to single crystals of a defect spinel phase after 48 hours at 1287$^\circ$C in air; normal disordered Panrimali sapphirine remains unchanged by similar heat treatment.

SAPPHIRINE occurs at Mautia Hill, Tanganyika, in enstatite-hornblende-sapphirine assemblages situated between bands of dolomite marble and quartz-yoderite-kyanite-talc schist (McKie, 1959) in poorly exposed ground just north of the crest of the hill. The sapphirine-bearing rocks, which are associated with högbomite-bearing enstatite-hornblende-chlorite-dolomite assemblages (McKie, 1963), have not yet been studied in detail in the field and it is the purpose of the present communication merely to describe the occurrence there of a sapphirine, remarkable for its yellow colour and for the evidence its X-ray diffraction pattern provides of partial ordering of Mg$^{2+}$ and Al$^{3+}$.

The limiting refractive indices of Mautia sapphirine, $\alpha 1.725 \pm 0.002$ and $\gamma 1.732 \pm 0.002$, are above the range commonly reported (Sørensen, 1955). The optic axial angle, determined on the universal stage as
2V_\gamma 66^\circ, with occasional values as high as 80^\circ, is anomalous. Sapphirine is usually optically negative with 2V_\alpha varying from 50^\circ to 'large'; Segnit (1957) alone has described optically positive sapphirine, with 2V_\gamma \sim 80^\circ, from Mawson Station, Antarctica. The new data extend the range of observed optic axial angles to 50^\circ \leq 2V_\alpha \leq 114^\circ. The extent of the variation is reminiscent of that in cordierite, where 2V_\alpha may lie between 39^\circ and 99^\circ, and is dependent on thermal history and composition. The orientation of the indicatrix, obtained by Laue photography of a crystal whose principal vibration directions had been marked by attached glass fibres, is normal with \beta || [010] and \gamma \wedge [001] \sim 7^\circ. The mineral appears strongly coloured in thin-section, is pleochroic with \alpha pale yellow, \beta pale lime-green, \gamma pale pinkish orange, and is highly dispersive. All sapphines described hitherto have had pleochroic schemes in the blue range. The colour of the Mautia material made its identification difficult; it was shown to be sapphirine by the identity of its X-ray powder pattern with that of sapphirine from Madura, Madras, by its unit-cell dimensions determined from single crystal X-ray data, and by its chemical composition. Occasional grains display polysynthetic twinning on \{100\}. Poor \{010\} and \{100\} cleavages are observable.

The unit-cell dimensions of sapphirine, originally determined by Gossner and Mussgnug (1928) by an unreliable method, have recently been redetermined by Dr. S. G. Fleet\(^1\) as a 9.77 Å, b 14.54 Å, c 10.06 Å, (all \pm 0.1 %), and \beta 100° 20' (\pm 2'). Fleet's observations were made on a pale blue crystal from Fiskenaesset, West Greenland, and lead to the space group \textit{P2}_1/n (in a non-standard orientation). Contemporaneous with Fleet's redetermination is that of Kuzel (1961), a 11.26 Å, b 14.46 Å, c 9.95 Å, \beta 125° 20', space group \textit{P2}_1/c. The Kuzel cell is derived from the Fleet cell by the matrix \{101/010/001\}. The Fleet cell is preferred and will be used in the sequel because it has a smaller \beta angle, as required by convention, and the systematic non-space-group absences characteristic of sapphirine obey simpler rules.

The X-ray diffraction pattern of Mautia sapphirine is anomalous in that it contains diffuse streaks parallel to \textit{b}*, on which lie diffuse intensity maxima with integral values of the index \textit{k} and sharp intensity maxima with fractional \textit{k} indices such that \textit{k} = (2\textit{n}+1)/2 in terms of the Fleet unit-cell. The length of the \textit{b}-axis must therefore be doubled

\(^1\) I am indebted to Dr. S. G. Fleet for permission to quote unit-cell dimensions, systematic absences, and preliminary structural conclusions from his unpublished Ph.D. thesis (1962, deposited in the University Library, Cambridge).
to give integral indices to all the sharp reflexions of Mautia sapphirine, the approximately measured unit-cell dimensions of which become $a \ 9.85 \ \text{Å,} \ b \ 28.6 \ \text{Å,} \ c \ 9.96 \ \text{Å,} \ \beta \ 110^\circ$. The space group remains $P2_1/n$. The reflexions observed on the diffraction pattern are assigned to various types of combination of indices in table I. Six types of sharp reflexion ($S$-reflexions) and two types of diffuse intensity maximum ($D$-reflexions) occur. The $S$-reflexions all obey the condition $k + l = 2n$, and those not on diffuse streaks the further simultaneous condition $k + 1 = 2n$.

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<tr>
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<td>$2n + 1$ (on streaks)</td>
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<table>
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<th>Type</th>
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<td>$D(7)$</td>
<td>$2n + 1$</td>
<td>$2n$</td>
<td>$2n + 1$</td>
</tr>
<tr>
<td>$D(8)$</td>
<td>$2n$</td>
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</tr>
</tbody>
</table>

$k = 2n$, $h + \frac{1}{2}k + \frac{1}{2}l = 2n$. The $D$-reflexions all obey the simultaneous condition $k = 2n$, $l = 2n + 1$ (i.e. $k + l = 2n + 1$). The diffuse streaks parallel to $b^*$ thus pass only through reciprocal lattice points with $l = 2n + 1$. The strongest streaks have $h = 2n + 1$, and join $S(3)$ and $D(7)$ reflexions; weaker streaks, with $h = 2n$, join $S(6)$ and $D(8)$ reflexions.

The diffraction pattern of normal sapphirine contains only sharp reflexions of types (1), (2), (4), (5), (7), and (8) (table I). In Mautia sapphirine the reflexions of types (7) and (8) have become diffuse, especially in the $b^*$ direction in reciprocal space, and new sharp reflexions of types (3) and (6) have appeared, requiring doubling of the $b$-axis. The Mautia material may be regarded as intermediate in structural state between normal sapphirine and a sapphirine exhibiting only sharp reflexions of types (1), (2), (3), (4), (5), and (6). Such an extreme structural state would have a unit-cell differing from that of normal sapphirine in being $A$-centred ($k + l = 2n$ for permitted reflexions) and in having $b$ doubled.

The salient features of the diffraction pattern of Mautia sapphirine
resemble those well known in hexagonal cobalt inverted from the high-
temperature cubic structure (summarized by Wilson, 1949, pp. 67–74). There reflexions are of three types: sharp reflexions with \( h-k = 3n, \ l = 2n \); reflexions with \( h-k \neq 3n, l = 2n \), drawn out into long streaks parallel to \( c^* \); and reflexions with \( h-k \neq 3n, l = 2n+1 \), much less diffuse. The diffuseness of the reflexions of the second and third types has been interpreted in detail as a consequence of mistakes in the stacking of (0001) layers of cobalt atoms, the mistakes being due to imperfect annealing of the low-temperature hexagonal polymorph. Some degree of resemblance also exists between the mutual relationship of the diffraction patterns of normal sapphirine, Mautia sapphirine, and the hypothetical extreme structural state and that of the diffraction patterns of the high-albite, body-centred anorthite, transitional anorthite, and primitive anorthite structures in the composition range \( \text{An}_{100} \) to \( \text{An}_{10/7} \) (Gay, 1953, 1954; Laves and Goldsmith, 1954). The size of the high-albite unit-cell, which produces only type \((a)\) reflexions (nomenclature of Gay, 1953), is increased by the appearance of diffuse \((b)\) reflexions, which become sharp in body-centred anorthite; in transitional anorthite diffuse \((c)\) reflexions appear, which become sharp and are accompanied by sharp \((d)\) reflexions in primitive anorthite. The structural sequence high-albite, body-centred anorthite, transitional anorthite, primitive anorthite, corresponds, broadly speaking, to decreasing temperatures of stability, or to increasing substitution of \( \text{CaAl} \) for \( \text{NaSi} \). It may be interpreted in general terms as an ordering sequence in that the high albite structure has a random distribution of \( \text{Al} \) and \( \text{Si} \) on tetrahedral sites, while the primitive anorthite structure is highly ordered; the body-centred and transitional anorthite structures appear to have ordered \( \text{Si-Al} \) arrangements and at least partial disorder in the distribution of the large cations (see, for instance, Bown and Gay, 1958).

The structure of sapphirine is not known, but Fleet’s Patterson syntheses show that the oxygen ions are very nearly in cubic close packing, the close packed unit being oriented such that \([10\overline{1}]\) is parallel to an edge of the close-packed oxygen cube and \([101]\) and \([010]\) are each parallel to diagonals of the cube face perpendicular to \([10\overline{1}]\). The small cations, \( \text{Si}^{4+} \) and possibly \( \text{Al}^{3+} \), would then be expected to lie in fourfold interstices and the large cations, \( \text{Mg}^{2+} \) and possibly \( \text{Al}^{3+} \), in sixfold interstices between the close-packed oxygen sheets. The possibility of an ordered or a random distribution arises for \( \text{Si}^{4+} \) and \( \text{Al}^{3+} \) on the fourfold sites and for \( \text{Mg}^{2+} \) and \( \text{Al}^{3+} \) on the sixfold sites. If the analogy with cobalt and the calcic plagioclase structures is accepted,
it follows that normal sapphirine would have a disordered structure, Mautia sapphirine a partially ordered structure, and that the hypothetical extreme structural state would be highly ordered. Ordering on fourfold and on sixfold sites cannot be distinguished in the absence of a structure determination. The special features of the diffraction pattern of Mautia (partially ordered) sapphirine do, however, seem more consistent with the extensive displacement of oxygen ions produced in MgAl ordering, as in yoderite, (McKie, 1959; Fleet and Megaw, 1962), than with the effects of AlSi ordering, as in albite. Why MgAl ordering should produce such different diffraction effects in the partially ordered state in Mautia sapphirine and in low-yoderite cannot be explained at this stage.

If the conclusions of the preceding paragraphs are valid, sapphirine must have crystallized at Mautia Hill either below the lower temperature limit of stability of normal (disordered) sapphirine, or it must have crystallized within the field of the disordered structural state and then been cooled slowly enough to produce good annealing within the field of partially ordered or ordered sapphirine. The choice is qualitatively the same as for the formation of low-yoderite but in the case of sapphirine no temperature can yet be attached to the order-disorder transformation although some preliminary dry heating experiments have been made.

Successive hearings in air of a single crystal of Mautia sapphirine contained in a carefully cleaned platinum microcrucible at 896°C for 24 h, at 1000°C for 24 h, at 1100°C for 24 h, and at 1200°C for 40 h produced no apparent change in its diffraction pattern, recorded as a b-axis oscillation photograph at room temperature after each period of heating. Further heating at 1287°C for 48 h transformed the single crystal of partially ordered sapphirine into a single crystal of a spinel phase. The same change was observed in another crystal heated at 1287°C for 65 h. The sapphirine diad symmetry axis persists as one of the spinel diads (110). The spinel phase is pale green, isotropic, and untwinned, with sharp X-ray reflexions which display only slight θ-streaking. The possibility of a significant change in composition produced by contamination was dismissed after observation of the transformation in a crystal selected from the exposed surface of a coarse powder heated in a platinum dish at 1395°C in air for 48 h. A diffractometer trace taken with Cu-Kα radiation over the range θ 15° to 20° shows progressive weakening of all sapphire peaks and development of 220 and 311 spinel peaks as the transformation proceeds at 1395°C.
in air. In contrast the diffractometer trace of a sample of normal disordered sapphirine from Panrimali, India, (78125) exhibits no change with similar heat treatment.

The transformation of partially ordered, but not of disordered, sapphirine to a, presumably defect, spinel phase at temperatures in excess of about 1240 ± 40° C points to the greater similarity of the ordered structure to the spinel structure. The production of single crystals rather than of polycrystalline aggregates indicates further that the two structures can differ only in detail. A more thorough study of the transformation on a high-temperature oscillation camera is in progress; reversal has not yet been achieved.

In order to assess the normality of the supposedly disordered structural state of sapphirine, crystals from a number of localities were examined by a-axis or c-axis oscillation photographs. Two crystals were examined from each occurrence. In the following list University of Cambridge Mineralogy Museum numbers are given in brackets and references are cited only where the occurrence is not mentioned in Sørensen's (1955) review of sapphirine parageneses:

Sapphirine–hornblende–phlogopite gneiss, Fiskenaesset, West Greenland (70921); sapphirine–spinel–plagioclase–hornblende–hypersthene gneiss, Sukkertoppen, West Greenland (19531); sapphirine–hypersthene–spinel rock, Hill Tracts, Vizagapatam, India (78123); cordierite–plagioclase–sapphirine–hypersthene gneiss, Ganguvarpatti, Madura, India (54886); sapphirine–sillimanite–cordierite rock, Panrimali, Madura, India (78125); sapphirine–spinel–corundum–titanomagnetite rock, Cortlandt, New York; ilmenite–rutile–sapphirine mass in anorthosite, St. Urbain, Quebec (193830); enstatite–sapphirine–cordierite rock, Mawson Station, MacRobertson Land, Antarctica (88193) (Segnit, 1957).

In none of these eight specimens were diffuse streaks parallel to b* or reflexions indicative of doubling of b seen on the oscillation photographs. Crystals from the dozen or so other known sapphirine localities have yet to be examined. It is, however, already clear that the peculiarities of the diffraction patterns of the ten crystals of sapphirine examined from Mautia Hill are unusual, if not unique.

The chemical composition of the Mautia sapphirine determined by Mr. J. H. Scoon is set down in column 1 of table II, and the contents of the asymmetric structural unit, equivalent to 20 O²⁻ are shown in column 1a. The mineral is characterized by relatively high contents of Mg²⁺, Mn²⁺, and Fe³⁺, although the total Fe content is not abnormal,
and it may be seen to lie clearly within the range of sapphirine plotted
in the (Mg,Fe)O-(Al,Fe)₂O₃-SiO₂ system on fig. 1. It most closely
resembles in composition two sapphrines from the sakenites of Madag-
agascar (Lacroix and de Gramont, 1921; Lacroix, 1940). The anomalous
yellow colour may be related to the high oxidation ratio, O.R. =
100Fe³⁺/(Fe³⁺ + Fe²⁺) = 66. If the pale blue colour normal in sapphi-
rine is due to the presence of the chromophore group Fe³⁺-O-Fe²⁺ at

Table II. Chemical composition of sapphirine and associated minerals from Mautia
Hill, Tanganyika

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<tr>
<th></th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>1a</th>
<th>2a</th>
<th>3a</th>
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<td>51.24</td>
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<td>99.84</td>
<td>O²⁻</td>
<td>20.000</td>
<td>24.000</td>
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1. Sapphirine, JH2800.
2. Enstatite, JH2800.
3. Hornblende, JH2800.

1a. Quarter-unit-cell contents in sapphirine calculated to 20 oxygen anions from
column 1.
2a. Half-unit-cell contents in enstatite calculated to 24 oxygen anions from
column 2.
3a. Half-unit-cell contents in hornblende calculated to 24 anions from column 3.

low total iron concentration, then sapphirine may be yellow when the
Fe³⁺ concentration is low and the Fe²⁺ concentration much lower. However, the oxidation ratio of Mautia sapphirine is not greatly in
excess of that of pale blue sapphirine from Fiskenaesset (O.R. 56,
Boeggild, 1954), and from Sakena (O.R. 57, Lacroix, 1940). An alter-
native possibility is that the chromophore group is Fe³⁺-O-Mn²⁺
and that the yellow colour is to be attributed to high manganese con-
centration in the presence of a high oxidation ratio.

Sapphirine occurs at Mautia Hill in rocks composed essentially of
course intergrown anhedral grains of enstatite, hornblende, and sapphi-
rine with abundant accessory pseudobrookite and hematite, and
occasional patches of dolomite. Certain of the sapphirine-bearing rocks have an interstitial matrix of fine-grained talc, which encloses oriented relics of each of the three principal minerals and is apparently derived alike from enstatite, hornblende, and sapphirine by retrogressive local

Fig. 1. Representation of sapphirine compositions in the system \((\text{Mg,Fe})_2\text{O}_3-(\text{Al,Fe})_2\text{Si}_2\text{O}_5-\text{SiO}_2\). Circles: Sapphirine, ○ Mautia (table II). ○ 1 Fiskenaesset, 2 Mawson, 3 Sakena, 4 Val Codera, 5 Blinkwater, 6 Dangin, 7 Ganguvarpatti (1–7 quoted from Deer, Howie, and Zussman, 1962, pp. 178–9), 8 Madagascar (Lacroix and de Gramont, 1921), 9 St. Urbain, Quebec (Warren, 1912), 10 synthetic sapphirine \(\text{Mg}_2\text{Al}_2\text{Si}_2\text{O}_8\) (Foster, 1950). □ Enstatite, Mautia (Table II). △ Hornblende, Mautia (Table II). En, end-member enstatite. Py, pyrope. Yd, yoderite. Cd, cordierite. Sp, spinel. Solid and broken lines indicate the sapphirine-bearing assemblage at Mautia Hill in terms of this system.

metasomatism; coarser, apparently primary or recrystallized talc also occurs in some specimens. Minor amounts of sapphirine are present in the neighbouring högbomite-bearing enstatite–hornblende–chlorite assemblages. The occurrence of högbomite and pseudobrookite in neighbouring highly oxidized assemblages is notable. The enstatite and hornblende of the rock from which the analysed sapphirine was separated have been analysed by Mr. J. H. Scoon; their compositions
ORDER–DISORDER IN SAPPHIRINE

are set down in columns 2 and 3 of table II and their cationic proportions calculated to 24 anions are shown in columns 2a and 3a respectively.

The enstatite is highly magnesian, oxidized, and aluminous. Its composition may be expressed in terms of $100 \text{Mg}^{2+}/(\text{Mg}^{2+} + \text{Fe}^{2+} + \text{Fe}^{3+} + \text{Mn}^{2+})$ as $\text{En}_{98}$. Its alumina content of 4-15 %, although not extreme, falls just within the range characteristic of high-pressure rocks such as those of the granulite facies; the alumina content of the host rock evaluated approximately by modal analysis, is 16 %. No quantitative conclusions about pressure and temperature of crystallization can, however, yet be drawn from the alumina content of ortho-pyroxenes (Boyd and England, 1960). The $R^{2+}/\text{Si}$ ratio is close to unity (1-003) as would be expected for an orthopyroxene in a rock that is neither markedly undersaturated nor contains free silica.

The hornblende also is low in iron, highly oxidized, and markedly aluminous. The occupation of the fourfold sites is close to Si$_7$Al. The sixfold sites are filled and clearly a small amount of Mg must be associated with Ca and the small Na content on eightfold sites.

| Be Ga Cr V Mo Li Ni Co Se Zr Cu Yt La Sr Pb Ba Rb |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Sapphirine... | 22 | 100 | 340 | --- | --- | 3 | 100 | 45 | --- | 60 | 22 | --- | --- | --- | 12 |
| Enstatite... | 2 | 2 | 8 | --- | --- | --- | 45 | 45 | --- | 100 | --- | --- | --- | --- | --- |
| Hornblende... | --- | 2 | 45 | 22 | --- | --- | 4 | 55 | 18 | 42 | --- | 10 | 100 | 12 | 55 | 450 |

Concentrations are given in p.p.m. A dash indicates a concentration below the limit of sensitivity. No other elements were sought.

The trace element content of the three analysed minerals, very kindly determined by optical spectrography for me by Dr. S. R. Nockolds, F.R.S., is shown in table III. Sapphirine, for which no comparable data are available, appears to be strongly enriched in Be, Ga, Cr, and Zr relative to the other two minerals.

No description of a sapphirine–enstatite–hornblende assemblage has appeared hitherto. Sapphirine is associated with enstatite, bronzite, or hypersthene in a number of occurrences, Fiskenaesset, Vizagapatam, Ganguvarpatti, Val Codera, Stallen, and Mawson (see the review of Sorensen, 1955, and, for Mawson, Segnit, 1957); in most of these, and, indeed, in most of its occurrences, sapphirine forms reaction rims about spinel or corundum. At Mautia there is no evidence of the formation of sapphirine from either spinel or corundum, and it appears to have crystallized in equilibrium with enstatite and hornblende. Sapphirine–amphibole parageneses are not uncommon; at Fiskenaesset the
amphiboles are gedrite and pargasite (Vogt, 1947), at Sukkertoppen and Ol.1 in Greenland and at Stallen hornblende, at Sakena edenite, and at Ganguvarpatti anthophyllite (see Sørensen, 1955). At most, if not all, of these occurrences it is doubtful whether sapphirine is in equilibrium with the amphibole. The presence of hornblende may be due to the moderate Ca content and relatively low Al content of the rock so that neither gedrite nor anorthite-bearing assemblages would be stable; sapphirine is the only essentially aluminous major constituent.

No experimental data on the stability of sapphirine-enstatite-amphibole assemblages, or, indeed, of any of the principal sapphirine parageneses are available. Sørensen (1955) suggests that sapphirine only forms at sites of local Al concentration under water-deficient conditions, but that would seem inapplicable at Mautia, where sapphirine does not apparently form by reaction of spinel. Turner (1958) regards hypersthene-spinel-sapphirine as a rare silica-poor assemblage of the hornblende granulite subfacies, the field of which he estimates at about 700°C and $p_{H_2O}$ in excess of about 7000 bars. Whatever may have been the physical conditions of formation of sapphirine at Mautia, it evidently crystallized slowly enough, or was subsequently annealed sufficiently slowly, to produce a partially ordered structure in contrast to the disordered structure of spinel-derived sapphirine.

The enstatite-hornblende-sapphirine and associated enstatite-hornblende-chlorite-hög bomite-dolomite rocks have chemical compositions qualitatively consistent with their being the products of reaction between dolomite-marble and the adjacent highly magnesian yoderite-bearing schist. The physical conditions under which the observed phase assemblages were generated are quantitatively uncertain, but apparently not dissimilar to those suggested by McKie (1959) for the formation of yoderite.

In conclusion it may be apposite to comment on the redox potential during metamorphic recrystallization of the sapphirine-bearing assemblage. The oxidation ratios of the constituent principal phases are 66, 88, and 83 for sapphirine, enstatite, and hornblende respectively. The relatively low total iron concentrations, 5.47 %, 1.41 %, and 2.89 % respectively in sapphirine, enstatite, and hornblende, make it improbable, however, that the uncommonly high state of oxidation was the controlling factor in the generation of this unique phase assemblage.

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