

## New Locality, Formula, and Proposed Structure for Reyerite

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### Abstract

Reyerite, a rare hydrated calcium silicate, has been discovered in chlorite-containing amygdules in a diabase dike in Rawlings quarry, Brunswick County, Virginia, that has been subjected to an episode of low-grade regional metamorphism. Microprobe, X-ray, optical, and infrared analyses of the Virginia specimen and of reyerite from the type locality at Niakornak, Greenland, show that they are remarkably similar (space group  $P\bar{3}$  or  $P3$ , according to Cann, 1965).

Virginia reyerite:



Greenland reyerite:



Reyerite is micaceous in appearance and the structure proposed for the  $(T_2O_5)_\infty$  sheet of corner-sharing tetrahedra is a modified mica sheet with every other  $T_6O_{18}$  ring rotated  $10^\circ$  clockwise about the lattice points in a multiple mica unit cell containing  $4(T_2O_5)$  groups. The six-fold rings between the rotated rings become distorted, and the most likely configuration is one in which every other tetrahedron points up and down normal to the sheet. There are three tetrahedral sheets stacked normal to  $c$  in such a way as to provide (1) distorted octahedral sites for Ca, small amounts of which may be included with the alkalis, and (2) hexagonal columns parallel to the  $c$  axis for zeolitic water.

Structural and chemical similarities of reyerite to gyrolite and truscottite are noted, but optical properties, infra-red spectra, and lattice parameters indicate that the three are most certainly distinct mineral species.

### Introduction

The rare mineral reyerite, a hydrated calcium silicate closely related to gyrolite and truscottite, has recently been discovered in a diabase dike in the Rawlings quarry, Brunswick County, Virginia. The lattice parameters, optical properties, composition, and mode of occurrence of this specimen closely match those of reyerite from the two previously reported localities.

Reyerite was first collected by Giesecke at Niakornak, Greenland, in 1811. It has been shown to be distinct from gyrolite (Mackay and Taylor, 1953), but its relationship to truscottite is less clear. Truscottite and reyerite were thought to be distinct minerals until Strunz and Micheelsen (1958) claimed to equate the two. Chalmers *et al* (1964) indicated that on the basis of optical properties, X-ray diffraction data, and especially infrared absorption spectra, the two minerals probably should remain as separate

species (*contra* Meyer and Jaunarajs, 1961). Cann (1965), assuming that reyerite and truscottite are distinct minerals, reported a second occurrence of reyerite at 'S Airde Beinn, Isle of Mull, Scotland.

Reyerite from Virginia was identified from its powder diffraction pattern; its similarity to that from Greenland and Scotland has been verified by microprobe analysis, lattice parameter refinement, infrared spectroscopy, and optical measurements. The U.S. National Museum kindly provided a small flake of reyerite from Greenland (Roebbling Collection No. 4016) for our use. The  $d$ -spacings for both the Virginia and the Greenland reyerites were measured by diffractometer using  $\text{CuK}\alpha$  radiation and silicon metal as an internal standard. Because the micaceous flakes were highly oriented in our powder mounts, the relative intensities differ greatly from those given by Chalmers *et al* (1964) for reyerite, but the  $d$ -spacings agree very closely. Lattice parameters

were refined using the program of Evans, Appleman, and Handwerker (1963), assuming space group  $P\bar{3}$  as deduced by Chalmers *et al* (1964) and Cann (1965) for their specimens.

### Composition and Physical Properties of Reyerite

The reyerites from Virginia and Greenland were analyzed on an ARL-SEM microprobe at VPISU using synthetic anorthite glass and a natural alkali feldspar (Smith and Ribbe, 1966, specimen 5748) as standards. Data were reduced using the Rucklidge and Gasparrini EMPADR VII program and the results are listed in Table 1. No transition metals were detected in these specimens.

Following Chalmers *et al* (1964) and consistent with the micaceous appearance of reyerite, we have normalized the formulas to 24 tetrahedral cations (Si + Al) and 60 oxygens, leading to a  $(T_2O_5)_\infty$  formula for the polygonized sheet of  $(Si,Al)_4$  tetrahedra. Hydroxyl content was determined arbitrarily by adding sufficient  $(OH)^-$  to formally balance the positive charges in excess of 120 *e.s.u.* The values are  $5.2(OH)^-$  and  $4.6(OH)^-$  for the Virginia and Greenland specimens, respectively. Based on dehydration studies, Chalmers *et al* chose  $5(OH)^-$  as the most likely value for the Greenland reyerite. Because we had insufficient pure material to determine

TABLE 1. Composition of reyerites from Virginia and Greenland

Oxide	Weight percent		Atom	Atoms per formula unit <sup>a</sup>	
	Virginia	Greenland		Virginia	Greenland
SiO <sub>2</sub>	53.4	54.7	Si	21.7	21.9
Al <sub>2</sub> O <sub>3</sub>	4.7	4.4	Al	2.3	2.1
CaO	31.8	31.6	Ca	13.9	13.6
Na <sub>2</sub> O	1.5	3.5	Na	1.1	2.7
K <sub>2</sub> O	<u>5.2</u>	<u>2.2</u>	K	2.7	1.1
Totals	96.6	96.4			
H <sub>2</sub> O <sup>b</sup>	3.4	3.6			

<sup>a</sup> Normalized to 24 (Si + Al)

<sup>b</sup> By difference from 100%. Some of the  $-400^\circ\text{C}$  water is likely to have been driven off by the heat of the electron beam. In dehydration experiments Chalmers *et al.* (1964) found that 3.4 to 4.2 wt. % H<sub>2</sub>O was lost below  $400^\circ\text{C}$  and approximately 2.6% (presumed to be  $(OH)^-$ ) above  $650^\circ\text{C}$ .

weight-loss curves or to measure the density of the Virginia reyerite, we follow Chalmers *et al* in assuming  $5H_2O$  to be correct for our specimens as well as theirs. Cann (1965) measured significantly more total H<sub>2</sub>O (10.4 wt percent) in the reyerite from Scotland (*cf.* footnote *b*, Table 1). He suggested that there is probably "... a substitution of  $4(OH)^-$  for  $SiO_4^{4-}$ ." The microprobe analyses indicate that no such substitution is necessary in reyerites from the other localities.

It is interesting to note that Al occupies approximately 10 percent of the tetrahedral sites in both the Virginia and Greenland reyerites. Comparison with similarly normalized wet chemical analyses of a reyerite from the same locality in Greenland and the one from Scotland show nearly identical substitutions, as do the gyrolites from Scotland and Italy (Table 2). Both the Virginia and the Greenland specimens contain  $\sim 3.8$  alkali atoms per formula unit, the former in the ratio 3 Na to 7 K, the latter 7 Na to 3 K; the numbers of Ca atoms are  $\sim 13.9$  and  $\sim 13.6$ , respectively and do not differ significantly within the estimated standard error of the analysis ( $\sim 2$  percent of the amount present). This chemical similarity is reflected in the statistically identical lattice parameters which, incidentally, are remarkably similar to those of the reyerite from Scotland whose non-tetrahedral cation and total H<sub>2</sub>O contents are substantially different from our specimens.

As seen in Table 2, the wet chemical analysis of the Greenland reyerite is in better agreement with the microprobe analysis than was indicated by the approximate formula,  $KCa_{14}Si_{24}O_{60}(OH)_5 \cdot 5H_2O$ , given by Chalmers *et al* in their abstract. Using their cell volume ( $1564.3\text{\AA}^3$ ), the approximate formula leads to a calculated density of 2.56 gm/cc, and their actual formula leads to a value of 2.62 gm/cc. The latter was judged to be in too poor agreement with their measured value of 2.54 gm/cc, although others have measured values as high as 2.578 gm/cc on the Greenland reyerite (Chalmers *et al.*, 1964). Using our observed composition and slightly larger cell volume ( $1574.6\text{\AA}^3$ ) for the Greenland reyerite (Table 2), we calculate a density of 2.59 gm/cc which is in quite reasonable agreement with the highest value reported by the earlier workers. It should be pointed out that the differences in lattice parameters for the two samples from Greenland are due to differences in the method of measurement: Chalmers *et al* used uncalibrated single-

TABLE 2. Comparison of Gyrolite, Reyerite, and Truscottite

Mineral, reference	Locality	Composition	Cell parameters <sup>a</sup>		
			a (Å)	c (Å)	Vol. (Å <sup>3</sup> )
<b>GYROLITE</b>					
Mackay & Taylor (1953)	Bombay	Ca <sub>16</sub> Si <sub>24</sub> O <sub>60</sub> (OH) <sub>8</sub> ·12H <sub>2</sub> O <sup>b</sup>	9.72(1)	6x22.13(5)	6x1810.7
Cann (1965)	Scotland	Ca <sub>16</sub> Si <sub>24</sub> O <sub>60</sub> (OH) <sub>8</sub> ·14H <sub>2</sub> O <sup>b</sup>	9.76(2)	3x22.26(4)	3x1836.3
REYERITE (normalized to 24(Si+Al), assuming sufficient (OH) <sup>-</sup> to balance charges)					
Chalmers et al. (1964)	Greenland	(Na <sub>,7</sub> K <sub>,3</sub> ) <sub>2,7</sub> Ca <sub>14,4</sub> (Si <sub>,9</sub> Al <sub>,1</sub> ) <sub>24</sub> O <sub>60</sub> (OH) <sub>5,1</sub> ·5H <sub>2</sub> O	9.74(1) <sup>c</sup>	19.04(2) <sup>c</sup>	1564.3 <sup>c</sup>
This study	Greenland	(Na <sub>,7</sub> K <sub>,3</sub> ) <sub>3,8</sub> Ca <sub>13,6</sub> (Si <sub>,9</sub> Al <sub>,1</sub> ) <sub>24</sub> O <sub>60</sub> (OH) <sub>4,6</sub> ·5H <sub>2</sub> O	9.765(3)	19.067(3)	1574.6
This study	Virginia	(Na <sub>,3</sub> K <sub>,7</sub> ) <sub>3,8</sub> Ca <sub>13,9</sub> (Si <sub>,9</sub> Al <sub>,1</sub> ) <sub>24</sub> O <sub>60</sub> (OH) <sub>5,2</sub> ·5H <sub>2</sub> O	9.764(3)	19.070(4)	1574.5
Cann (1965)	Scotland	(Na,K,Mg,Fe) <sub>2</sub> Ca <sub>14,2</sub> (Si <sub>,9</sub> Al <sub>,1</sub> ) <sub>24</sub> O <sub>60</sub> (OH) <sub>y</sub> ·xH <sub>2</sub> O	9.76(2)	19.10(2)	1575.7
<b>TRUSCOTTITE</b>					
Mackay & Taylor (1954)	Sumatra	Ca <sub>12</sub> Si <sub>24</sub> O <sub>60</sub> ·6H <sub>2</sub> O <sup>d</sup>	9.72(1)	18.71(3)	1527.7
Minato & Kato (1967)	Japan	Ca <sub>12</sub> Si <sub>24</sub> O <sub>60</sub> ·8H <sub>2</sub> O <sup>d</sup>	9.72	18.84	1541.5

<sup>a</sup> Estimated standard errors are in parentheses and refer to the last decimal place.

<sup>b</sup> Approximate, ideal formulas. It is difficult to reconcile these with the actual chemical analyses. Both show 5-10% Al for Si, as in reyerite. Cann's analysis, normalized to 24(Si+Al), is (Na,K,Mg,Fe)<sub>2</sub>Ca<sub>13</sub>(Si<sub>,9</sub>Al<sub>,1</sub>)<sub>24</sub> plus anions and water. It is interesting to note that gyrolite from Monte Biaena, Italy also contains 10% Al substituting for Si (Gottardi and Passaglia, 1968).

<sup>c</sup> These are the values given by Chalmers et al. A least-squares refinement of the *d*-spacings reported for their powder pattern gave values of *a* = 9.766(1)Å; *c* = 19.047(3)Å; and Vol. = 1573.4Å<sup>3</sup>, in rather good agreement with those determined in this study.

<sup>d</sup> Approximate formulas. Chemical analyses show alkali and transition metals and very minor Al. The truscottite from Japan contains 2.41 wt.% MnO.

crystal photographs to determine *a* and *c*, but using the *d*-spacings they reported for their powder pattern, we refined *a* and *c* and found them to be in very close agreement to our values (see Table 2, especially footnote *c*). There is extremely close agreement of cell volumes for the Greenland and Virginia reyerites, but the latter has a higher calculated density (2.62 gm/cc based on an assumed 5H<sub>2</sub>O) because it contains more K and Ca than the former.

Infrared spectra over the range 2.5 to 40 microns were recorded for both reyerites using KBr discs in a grating spectrophotometer. The position of the absorption bands are identical for the two samples but there are slight differences in absorption within the 12 to 15 micron range. The bands in this range are more clearly defined for the Virginia than for the Greenland reyerite (this study) but match closely those shown by Chalmers et al (1964, Fig. 3, p. 831) for material from Greenland.

As shown in Table 3, the optical properties of reyerite from Virginia are very similar to those previously described. All are uniaxial negative with a birefringence of 0.005, in contrast to uniaxial negative truscottites which have refractive indices *n<sub>e</sub>* ~1.53 and *n<sub>w</sub>* ~1.55 (Minato and Kato, 1967).

### Proposed Sheet Structure for Reyerite

The crystal structure of reyerite has not been determined, but it is nonetheless clear that the principal structural units are infinite sheets of corner-sharing (Si<sub>0,9</sub>Al<sub>0,1</sub>)O<sub>4</sub> tetrahedra parallel to (0001). Reyerite is chemically similar to gyrolite and truscottite (see Table 2) which are also hydrated calcium silicates. Mackay and Taylor (1953) indicated that gyrolite from Bombay is hexagonal *P*6<sub>1</sub> and that there is considerable stacking disorder along the *c* = 6 × 22.13 Å axis. Cann's (1965) gyrolite from Mull, although optically biaxial, is strongly pseudo-hexagonal showing similar disorder along the *c* = 3 × 22.26 Å axis. According to both Cann and Chalmers et al (1964), reyerite does not exhibit significant stacking disorder; its *c* axis is 19.07 Å, some 3Å shorter than the more hydrous gyrolite.

TABLE 3. Refractive Indices of Reyerite

Locality	Reference	<i>n<sub>w</sub></i>	<i>n<sub>e</sub></i>
Greenland	Bøggild (1908)	1.5645	1.5590
Greenland	Chalmers et al. (1964)	1.563	1.558
Scotland	Cann (1965)	1.568	1.563
Virginia	This study	1.568	1.563

The space group of reyerite is  $P\bar{3}$  (Cann, 1965), not "monoclinic, pseudo-trigonal" as suggested by Meyer and Jaunarajs (1961), and the polygonization and stacking of the tetrahedral sheets must be consistent with this symmetry; *i.e.*, the primary units must be 3-, 6-, 9- or 12-fold rings, although 9- and 12-fold rings are probably disallowed by the relatively small (9.76 Å) *a*-axis repeat.

Mackay and Taylor (1953) first discussed the nature of the tetrahedral sheets in gyrolite, which has an *a* axis repeat very similar to that in reyerite. They observed that the 5.2 Å simple hexagonal unit of a mica-like tetrahedral sheet contains one  $\text{Si}_2\text{O}_5$  group, but that a multiple cell containing three  $\text{Si}_2\text{O}_5$  groups can be chosen with a 9.2 Å *a* axis. Since it would obviously require severe distortion of the interatomic angles and an impossible extension of the Si-O bonds to  $>1.7\text{Å}$  to stretch the *a* axis to 9.76 Å, they abandoned their search for a model.

Strunz and Micheelsen (1958) suggested that zeophyllite and reyerite have similar sheet structures, but this seems unlikely because zeophyllite sheets have the formula  $(\text{Si}_3\text{O}_8)_\infty$ , not  $(\text{Si}_2\text{O}_5)_\infty$ . Meyer and Jaunarajs (1961), working with synthetic hydrous calcium silicates, claim a CaO/SiO<sub>2</sub> ratio of 0.5 for both "gyrolite" and "reyerite" (probably in fact truscottite). They hypothesized that their synthetic compounds consist of layers with trigonal symmetry stacked in an open manner to permit inter-layer H<sub>2</sub>O in gyrolite (*c* ~22 Å) and in a tighter manner in "reyerite" (*c* ~18.7 Å, probably truscottite). The results of the investigations of natural materials contradict their arguments: Table 2 shows the clear distinction between reyerite (Na + K + Ca ≈ 17; *c* ~19.1 Å) and truscottite (Ca ≈ 12; *c* ~18.7 Å). Differences in optical properties confirm the distinction, as mentioned earlier.

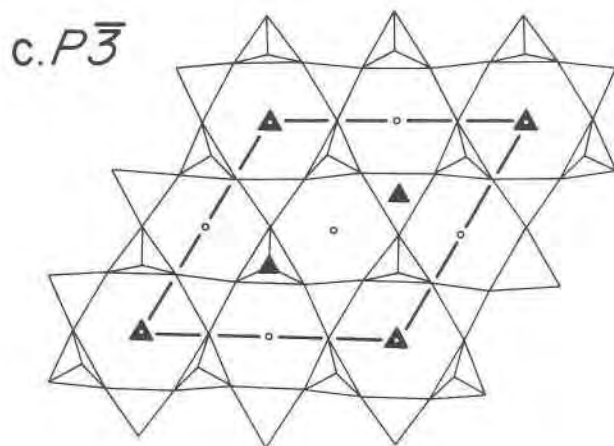
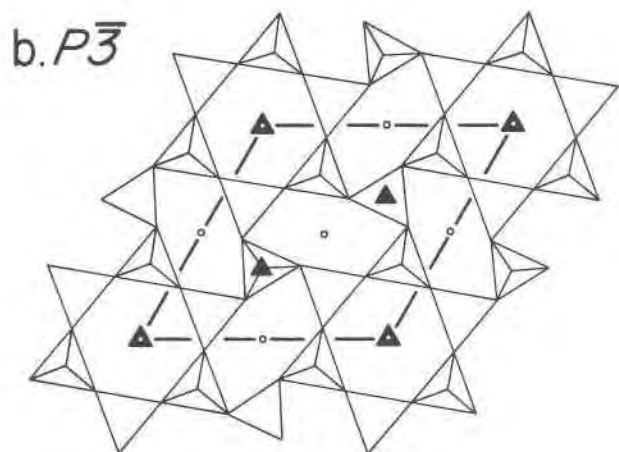
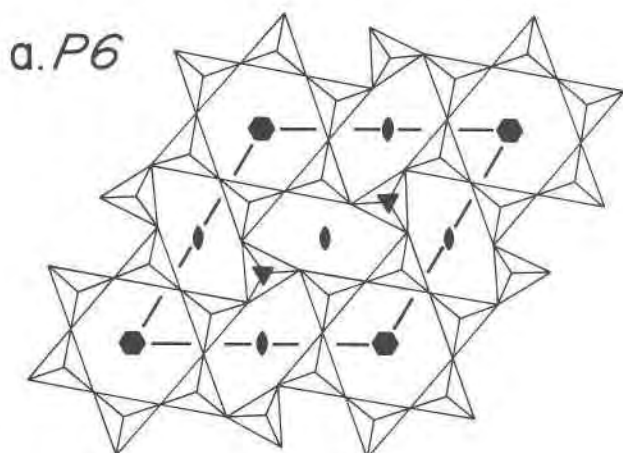
Chalmers *et al* (1964) hypothesized that the sheets in reyerite might be based on  $\text{Si}_6\text{O}_{18}$  rings because, when natural reyerite is dehydrated (above 650°C),  $\alpha\text{-CaSiO}_3$  is formed and its structure is based on  $\text{Si}_3\text{O}_9$  rings in an epitaxial orientation. Their Figure 5 (p. 835) shows the structural relationship, albeit not to scale, between the proposed  $\text{Si}_6\text{O}_{18}$  rings of reyerite and the  $\text{Si}_3\text{O}_9$  rings of  $\alpha\text{-CaSiO}_3$ .

We have chosen a multiple hexagonal cell of the mica sheet with *a* = 10.4 Å and four  $\text{Si}_2\text{O}_5$  groups, and instead of attempting to stretch bonds as Mackay and Taylor mentioned, we simply rotate the  $\text{Si}_6\text{O}_{18}$  rings at the lattice points 10° in a clockwise direction

to achieve a geometrically reasonable configuration. The result of this rotation is shown in Figure 1: every other hexagonal ring is distorted with the effect of shortening the *a* axis from 10.4 to 9.76 Å. If all the tetrahedra in the sheet point upwards, the sheet has symmetry consistent with space group  $P6$  (Fig. 1a). If, however, adjacent tetrahedra point in opposite directions, the symmetry of the sheet is  $P\bar{3}$  (Fig. 1b). A geometrically more regular polygonization of the sheet can be obtained by starting with a 10.4 Å cell as before and tilting the downward-pointing tetrahedra in the  $\text{Si}_6\text{O}_{18}$  rings down and the upward-pointing tetrahedra up by 15–20° from the plane of the rings (Fig. 1c) instead of rotating and distorting every other ring (as in Fig. 1b).

Inasmuch as there are 12  $\text{T}_2\text{O}_5$  groups per unit cell and 4 per sheet, there must be 3 sheets stacked along the *c* axis. And if reyerite is indeed centrosymmetric (*i.e.*  $P\bar{3}$ , not  $P3$ ), there must be two symmetrically non-equivalent sheets. It is assumed that the Al is disordered in the *T* sites within a given sheet, although with two different types of sheets all the Al may be disordered in one or the other. With the sorts of sheets suggested for reyerite, it is possible to contrive stacking sequences consistent with the symmetry that produce distorted octahedral inter-layer sites for the Ca atoms. It is likely that the 5 (ideally 6?) hydroxyls form part of the Ca coordination polyhedra, but it may be that only twelve of the calcium atoms are accommodated between the tetrahedral layers and that the remaining Ca plus non-integral and apparently variable amounts of K, Na and H<sub>2</sub>O are located in the hexagonal columns parallel to *c*. The oxygens in the  $(\text{Si}_{0.9}\text{Al}_{0.1})_6\text{O}_{18}$  rings are certainly charge-underbalanced (some more than others) and these would provide a sufficient field to hold the alkali atoms. The water is zeolitic in nature and is only weakly bonded, as evidenced by the ease with which preliminary dehydration occurs and the fact that the structure is unaltered in the process (Chalmers *et al*, 1964).

In summary it should be emphasized that until a crystal structure analysis is performed, the configuration of the sheets will remain hypothetical. It is in fact possible to build a geometrically plausible model for reyerite consisting of independent  $(\text{Si,Al})_{12}\text{O}_{30}$  groups bonded laterally into (0001) sheets by calcium atoms in irregular 7- or 9-fold coordination with oxygen and hydroxyl ions. Alkalies and water may be inter-layer or intracolumnar or both. The similarity in lattice parameters as well as composi-



tion indicate that the structure(s) of the sheet(s) in reyerite will be nearly the same as those for gyrolite and truscottite, which are nonetheless distinct mineral species. The sheet we proposed in Figure 1b differs only in polygonization from that proposed for triclinic okenite ( $\text{Ca}_9\text{Si}_{18}\text{O}_{45} \cdot 6\text{H}_2\text{O} \cdot 12\text{H}_2\text{O}$ ) by Mamedov and Belov (1958) and that observed in chemically similar tetragonal apophyllite ( $\text{K}_2\text{Ca}_8\text{Si}_{16}\text{O}_{40}\text{F}_2 \cdot 16\text{H}_2\text{O}$ ) by Taylor and Naray-Szabo (1931) (*cf* Bloss, 1971, Fig. 9–37).

#### Location and Paragenesis of the Virginia Reyerte

Reyerite occurs in amygdules at all known localities: in Greenland it is found within a tuff, in Scotland within the plateau basalts and in Virginia within a diabase dike. The Virginia locality is the abandoned Rawlings quarry south of the town of McKenney in Brunswick County that was once operated by the Southern Materials Company, Inc., for gneissic road aggregate. Specimens were collected from a narrow zone near the center of a vertical diabase dike that is approximately fourteen feet thick. At the present time slope wash conceals the collecting site at the southeast corner of the quarry.

The large dike has well developed chill zones at each contact and exhibits a gradational increase in grain size toward its center. Amygdule frequency and size as well as gross mineralogy change from the walls to the center of the intrusive (Clement, 1969). The primary minerals are plagioclase, augite, and olivine, with small amounts of apatite and opaques. Textures range from aphanitic prophyritic at the margins of the dike to intergranular to diabasic near the center. Secondary minerals include serpentine, localized concentrations of chlorite, and iddingsite that has almost entirely replaced olivine. Minerals found in the amygdules include reyerite, analcime, chlorite, calcite, pectolite, and traces of an unidentified radiating fibrous mineral, probably natrolite.

In the chill zone, nearly spherical amygdules are



FIG. 1. Proposed structures for the  $(\text{T}_2\text{O}_5)_\infty$  sheet in reyerite.  
 a. All tetrahedra pointing in the same direction; hexagonal rings at the lattice points rotated  $10^\circ$  clockwise; symmetry  $P6$ .  
 b. Every other tetrahedron pointing up and down; hexagonal rings at the lattice points rotated  $10^\circ$  clockwise; symmetry  $P\bar{3}$ .  
 c. Every other tetrahedron pointing up and down and tilted out of the plane of the sheet; hexagonal rings at the lattice points unrotated relative to a  $10.4\text{\AA}$  mica cell; symmetry  $P\bar{3}$ .

abundant and average 0.2 mm in diameter whereas near the center of the dike they are less common but larger, reaching approximately 10 mm in diameter. In all cases the amygdules are lined with chlorite. The small amygdules in the chill zones are either completely filled with chlorite or have a chlorite rim and a calcite core. Those in the central portion of the dike commonly have cores of analcite. As far as could be determined, reyerite-bearing amygdules are restricted to a zone approximately one foot thick near the center of the dike.

Within this zone reyerite occurs in two modes, either as a relatively pure core in a chlorite-rimmed amygdule or as a complex intergrowth with chlorite. In the first case the reyerite forms radiating aggregates of pearly white to pale green flakes. The greenish color is due to chlorite interleaved parallel to the perfect basal cleavage of the reyerite. In the second case the reyerite is poikilitic and contains numerous scattered intergrowths of chlorite that in thin section appear as rosettes and radiating crystal groups.

The close association of reyerite with other alteration minerals suggests that it formed subsequent to the emplacement of the dike. The Rawlings quarry dike and one other small dike in the nearby Dinwiddie quarry differ from all other exposed diabase dikes in the region in that they are amygdaloidal, contain (or contained) olivine, and are highly serpentinized and chloritized. Slickensides are common on many surfaces. The uniqueness of the two dikes was confirmed by Dr. P. Weigand (written communication, 1969) who indicated that of over two hundred diabase dikes studied from the eastern Piedmont of the Carolinas and southern Virginia, the Rawlings and the Dinwiddie dikes are the most metamorphosed. Kohler (1971) in a study of the alkali feldspars in the contact aureole surrounding the Dinwiddie dike concluded that the thermal effects of the intrusion have been obliterated by subsequent low grade regional metamorphism. The metamorphic origin for the Virginia reyerite is supported by the work of Cann (1965), who found the Scotland reyerite within the contact aureole of a volcanic plug. He postulated that it formed through the mild metamorphism of gyrolite, a mineral not yet identified in Virginia. It is concluded that the reyerite-bearing diabase dike in the Rawlings quarry has been subjected to an episode of low grade regional metamorphism.

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