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Amphibole and biotite  
from rapakivi

by Ahti Simonen and Atso Vormaa

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AHTI SIMONEN AND ATSO VORMA

WITH 8 FIGURES AND 8 TABLES IN TEXT

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## ABSTRACT

The petrographic characteristics of the rapakivi varieties occurring in the rapakivi massif of Wiborg (Viipuri) in southeastern Finland are described and the associations of the Fe-Mg-silicates reported. The occurrence, X-ray crystallography, chemical compositions and physical properties of rapakivi ferrohastingsite, grunerite and biotite are described. Extremely iron-rich varieties of ferrohastingsite and biotite, which crystallized from rapakivi magma, are the most common mafic minerals of the rapakivi. Grunerite is found in small quantities in certain dark-coloured varieties of the rapakivi and it is usually homoaxially intergrown with ferrohastingsite. The grunerite is the result of deuteric or hydrothermal reactions after the magmatic crystallization of the rapakivi. The cation distribution in the rapakivi Fe-Mg-silicates is described and discussed.

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## INTRODUCTION

Petrographic study of the specimens collected during the geological remapping of the broad rapakivi area in southeastern Finland (the so-called rapakivi massif of Wiborg (Viipuri) with the satellitic massifs of Suomenniemi and Mäntyharju) has shown that the Fe-Mg-silicates of the different rapakivi varieties consist of olivine, pyroxene, amphibole and biotite, and that their alteration products are iddingsite and chlorite. The mineralogy of the olivine from the Finnish rapakivi has been reported previously (Simonen, 1961 a) and now the present authors have undertaken to describe the amphibole and the biotite, which are the most common mafic mineral constituents of the rapakivi. Some mineralogical data on the amphibole and biotite in the rapakivi granites have already been reported by various authors (e.g., Sahama, 1947; Eskola, 1949; Savolahti, 1962; Sviridenko, 1967; Sobolev, 1947) and they point to extremely iron-rich varieties of Fe-Mg-silicates. To complement and increase the knowledge concerning the mineralogy of Finnish rapakivi, the authors will describe the occurrence, X-ray crystallography, chemical and physical data of the rapakivi amphibole and biotite.

## PETROGRAPHIC CHARACTERISTICS OF RAPAKIVI VARIETIES

The rapakivi massif in southeastern Finland represents an anorogenic pluton, whose emplacement into the stable platform took place about 1 650 million years ago. This broad rapakivi area is composed of many petrographic varieties of potash-rich granitic rocks. The various rapakivi granites differ from each other mainly in regard to textural features, but there are also slight differences between the principal varieties in chemical and mineralogical composition.

In the scope of the present study, it is important to note that potash feldspar, plagioclase and quartz are the main minerals contained in all the rapakivi varieties and only slight deviations occur in their relative contents. The most remarkable deviations between the different rapakivi varieties appear in the mineral associations of the Fe-Mg-silicates. Hornblende and biotite are the most common mafic mineral components of the rapakivi, in addition to which certain varieties contain olivine and pyroxene.

TABLE 1  
Distribution of the different rapakivi varieties in the Finnish part of the Wiborg (Viipuri)  
rapakivi massif

Rock type	%
wiborgite .....	76.2
dark-coloured wiborgite .....	4.9
pyterlite .....	6.1
porphyritic rapakivi granite .....	1.2
dark-coloured rapakivi granite .....	3.1
even-grained rapakivi granite .....	7.8
porphyry aplite .....	0.7
quartz porphyry and granite porphyry .....	< 0.1
aplite and pegmatite .....	< 0.1

The rock types mapped and their areal distribution in the Finnish part of the Wiborg (Viipuri) rapakivi massif are presented in Table 1. Some short notes on the petrographic characteristics of the principal rapakivi varieties follow:

Wiborgite, which shows a typical rapakivi texture, is the most abundant rock type of the rapakivi area. It is a coarse-grained porphyritic granite with ovoids of orthoclase surrounded by plagioclase mantles. Mantled ovoids are densely distributed. The main minerals are potash feldspar, plagioclase and quartz. Hornblende and biotite are the typical mafic silicates, and alteration products are iddingsite and chlorite. The chemical composition of a wiborgite is presented in Table 2, anal. 3.

The dark-coloured wiborgite has a sparse content of potash feldspar ovoids, which are mantled by plagioclase. In addition, this variety contains dark-coloured, zoned plagioclase as porphyritic grains. The main minerals of the dark-coloured wiborgite are the same as in the normal wiborgite, but in addition it contains olivine, iddingsite and grunerite. Furthermore, the plagioclase and hornblende contents are higher than those in normal wiborgite. The chemical composition of a dark-coloured wiborgite is given in Table 2, anal. 2.

The pyterlite is a coarse-grained porphyritic granite with ovoids of potash feldspar, but around the ovoids there are either no mantles of plagioclase or only very sporadic occurrences of them. The contents of potash feldspar and quartz are higher and that of plagioclase lower than in normal wiborgite. The most characteristic Fe-Mg-silicate is biotite, whose alteration product is chlorite. Hornblende is usually lacking. The chemical composition of a pyterlite is presented in Table 2, anal. 4.

The porphyritic rapakivi granite is characterized by an abundance of angular porphyritic grains of potash feldspar without mantles of plagioclase. The mineralogical as well as chemical composition of this variety is quite similar to that of the pyterlite. The chemical composition of a porphyritic rapakivi granite is given in Table 2, anal. 5.

The dark-coloured rapakivi granite is texturally an even-grained rock of medium or coarse composition. Some varieties contain, however, large ovoids of potash

feldspar. The green colour of the rock is due to the weed texture produced by the green hornblende. This green-coloured rapakivi variety has been called tirilite (*cf.*, Hackman, 1934). It contains abundant quartz, plagioclase and potash feldspar in highly variable proportions, and hornblende, biotite, olivine and pyroxene are the mafic silicates. The olivine is an iron-rich member of the forsterite-fayalite series and the forsterite content ranges from 2 to 8 mol. per cent (Simonen, 1961 a). Monoclinic pyroxene occurs sporadically. Hornblende is always present and in greater amounts than biotite. The alteration products of the aforementioned Fe-Mg-silicates are iddingsite, grunerite and chlorite. The mineralogical and chemical composition of the dark-coloured rapakivi is related to that of the dark-coloured wiborgite. The chemical composition is given in Table 2, anal. 1.

The even-grained rapakivi granite is medium- or coarse-grained, potash-rich granite. Usually red-coloured, it sometimes also has a greyish tint. The main minerals are potash feldspar, quartz and plagioclase. Biotite, altered partly into chlorite, is the main mafic mineral. Hornblende occurs only sporadically. The mineralogical as well as chemical composition of the even-grained rapakivi granite is similar to that of the pyterlite. The chemical composition may be seen in Table 2, anal. 6.

The porphyry aplite is a rare rapakivi variety that contains sparsely distributed, mantled or unmantled ovoids of potash feldspar in a fine-grained aplite-granitic matrix. The mineralogical and chemical composition of the porphyry aplite is similar to that of the pyterlite and even-grained rapakivi granite. The chemical composition is given in Table 2, anal. 7.

The dikes of quartz porphyry and granite porphyry cut the principal rapakivi varieties and the migmatitic country rocks of the rapakivi massif. The mineralogical and chemical composition of these rocks is similar to that of the even-grained rapakivi granite.

The aplites and pegmatites, which cut the principal rapakivi types, are both mineralogically and chemically related to the even-grained rapakivi granite.

The most marked differences between these rapakivi varieties appear in the textural features. The rapakivi granites are predominantly coarse-grained, porphyritic granites with large ovoids of potash feldspar commonly surrounded by plagioclase mantles (wiborgite type). In some varieties the plagioclase mantles around potash feldspar ovoids are almost totally lacking (pyterlite type). The ratio between the mantled and non-mantled ovoids varies greatly and texturally there is a gradual transition from the wiborgite to the pyterlite. The porphyritic granites with porphyritic, angular potash feldspar grains are likewise sporadically present and a gradual transition from this rock to the pyterlite is observed. The proportions of mantled and non-mantled and angular porphyritic potash feldspar grains in the coarse-grained, porphyritic varieties (wiborgite-pyterlite-porphyritic rapakivi granite) of the rapakivi in the Wiborg (Viipuri) massif are presented in the triangular diagram (Fig. 1). Based on 110 determinations made during the field work, this diagram indicates



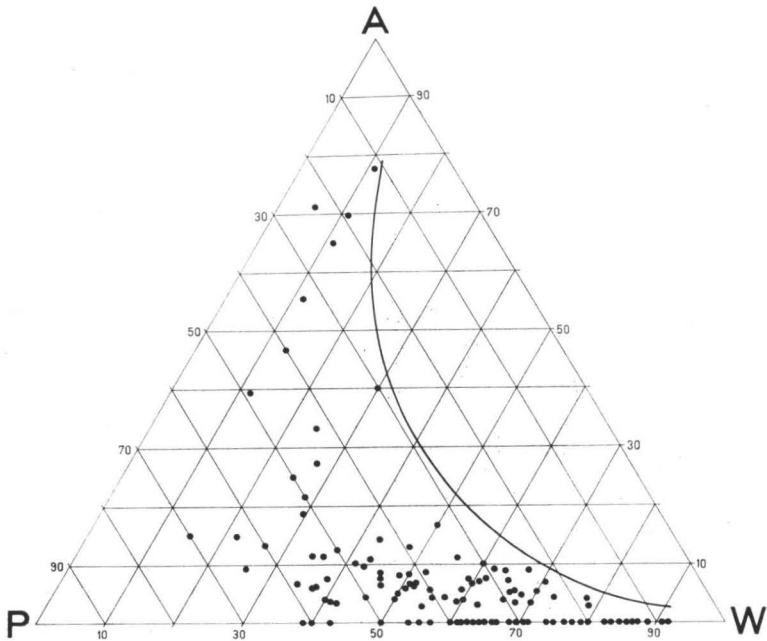


FIG. 1. Ratios between mantled, non-mantled and angular porphyritic potash feldspar grains in the rapakivi rock series, wiborgite-pyterlite-porphyrific rapakivi granite, in the Wiborg (Viipuri) rapakivi massif. *W*, potash feldspar ovoids mantled by plagioclase (wiborgite type); *P*, potash feldspar ovoids without plagioclase mantle (pyterlite type); *A*, angular, porphyritic potash feldspar grains (porphyritic rapakivi granite type).

that among the rapakivi varieties there are gradual transitions from the wiborgite via pyterlite into the porphyritic rapakivi granite with angular potash feldspar grains.

The chemical compositions of typical representatives of the different rapakivi varieties are presented in Table 2. All the rapakivi varieties are chemically characterized by high contents of silica and potassium, whereas the contents of lime and magnesia are low. The dark-coloured wiborgite and dark-coloured even-grained granite contain less silica and more FeO, MgO and CaO than other rapakivi varieties. Furthermore, the contents of FeO, MgO and CaO in the wiborgite are higher than those in the pyterlite. The pyterlite, porphyritic rapakivi granite, even-grained rapakivi granite, porphyry aplite and quartz porphyry are chemically closely related. The average chemical composition of the rapakivi has been determined by Sahama (1945), who has also done trace element determinations, which show that there are enrichments of fluorine, zirconium, rubidium and lead in the rapakivi granites.

TABLE 2

Chemical compositions of different rapakivi varieties in the Wiborg (Viipuri) rapakivi massif. Anal. by P. Ojanperä and A. Heikkinen. Fluorine determined by A. Heikkinen

	1	2	3	4	5	6	7
SiO <sub>2</sub> .....	63.72	67.11	68.88	76.69	76.20	74.87	74.38
TiO <sub>2</sub> .....	0.96	0.45	0.49	0.25	0.15	0.23	0.27
Al <sub>2</sub> O <sub>3</sub> .....	14.29	15.40	13.74	10.68	11.90	12.39	12.10
Fe <sub>2</sub> O <sub>3</sub> .....	1.92	0.89	1.02	0.96	0.27	0.25	0.75
FeO .....	5.74	3.42	3.62	1.93	1.54	1.89	1.89
MnO .....	0.13	0.05	0.06	0.03	0.02	0.02	0.03
MgO .....	0.69	0.42	0.47	0.17	0.02	0.24	0.31
CaO .....	3.28	3.16	1.92	1.01	1.16	0.82	0.78
Na <sub>2</sub> O .....	2.68	2.85	2.94	2.27	2.54	2.42	2.53
K <sub>2</sub> O .....	4.73	4.86	5.79	5.15	5.20	5.79	5.76
P <sub>2</sub> O <sub>5</sub> .....	0.38	0.15	0.14	0.04	0.02	0.03	0.04
H <sub>2</sub> O+ .....	0.64	0.66	0.75	0.47	0.41	0.47	0.56
H <sub>2</sub> O- .....	0.14	0.11	0.20	0.08	0.09	0.08	0.13
CO <sub>2</sub> .....	0.44	—	—	—	—	—	—
F .....	—	0.20	0.21	0.40	0.33	0.42	0.33
Li <sub>2</sub> O .....	—	0.03	0.06	0.02	0.04	0.05	0.03
Rb <sub>2</sub> O .....	—	0.04	0.04	0.05	0.13	0.06	0.06
—O = F <sub>2</sub> .....	99.74	99.80	100.33	100.20	100.02	100.03	99.95
	—	0.08	0.09	0.17	0.14	0.18	0.14
		99.72	100.24	100.03	99.88	99.85	99.81

1. Dark-coloured rapakivi granite (tirilite). Tullisenlampi, Lemi. Anal. P. Ojanperä.
2. Dark-coloured wiborgite. NW of Pyhäjärvi, Artjärvi. Anal. P. Ojanperä.
3. Wiborgite. Road cut, Lapinjärvi. Anal. P. Ojanperä.
4. Pyterlite. Sutela, Kotka. Anal. P. Ojanperä.
5. Porphyritic rapakivi granite. Verla, Iitti. Anal. A. Heikkinen. Li<sub>2</sub>O and Rb<sub>2</sub>O determined by P. Ojanperä.
6. Even-grained rapakivi granite. W of Kirkkojärvi, Myrskylä. Anal. P. Ojanperä.
7. Porphyry aplite. Porlammi, Lapinjärvi. Anal. P. Ojanperä.

TABLE 3

Fe-Mg-silicates of rapakivi varieties. The + sign marks minerals regularly present, (+) those occasionally present

Rock types	olivine	pyroxene	iddingsite	grunerite	hornblende	biotite	chlorite
dark-coloured rapakivi granite (tirilite) ..	+	(+)	+	(+)	+	+	+
dark-coloured wiborgite .....	+		+	+	+	+	+
wiborgite .....			(+)		+	+	+
pyterlite .....					(+)	+	+
porphyritic rapakivi granite .....						+	+
even-grained rapakivi granite .....					(+)	+	+
porphyry aplite .....					(+)	+	+
quartz porphyry and granite porphyry ..					(+)	+	+
aplite and pegmatite .....						+	+

The mineralogical composition of all the rapakivi varieties is characterized by the abundance of potash feldspar (mainly orthoclase), plagioclase (mainly oligoclase) and quartz. Furthermore, the potash feldspar is predominant over the plagioclase. The dark-coloured rapakivi granite and dark-coloured wiborgite contain, however, more plagioclase and less potash feldspar and quartz than do other rapakivi varieties. The mineral associations of the Fe-Mg-silicates found in the rapakivi varieties are presented in Table 3. Olivine, monoclinic pyroxene and grunerite have been found only in the dark-coloured varieties of rapakivi. Hornblende and biotite are the most common mafic minerals of the rapakivi and the mineral association hornblende-biotite is typical of the normal wiborgite, which is the most common type of rapakivi. The biotite is the main mafic mineral of the pyterlite and other highly silicic varieties of the rapakivi.

In the following chapters the mineralogical data are given for the rapakivi amphibole and biotite. The present study has shown that the rapakivi amphibole is represented both by hornblende and grunerite. Furthermore, the hornblende is seen to be an iron-rich variety corresponding to ferrohastingsite.

## FERROHASTINGSITE

### Occurrence

Ferrohastingsite is the main mafic mineral of many rapakivi varieties. It is most abundant in the dark-coloured varieties of rapakivi, which may contain as much as 10 to 15 per cent of ferrohastingsite. Moreover, ferrohastingsite is a typical mineral of normal wiborgite, but it occurs only occasionally in the most silicic rapakivi varieties. The Fe-Mg-silicates coexisting with ferrohastingsite are listed in Table 3.

### X-ray crystallography

The rapakivi ferrohastingsites, Nos. 1, 2, 3 and 5, in Table 4 were investigated by the Buerger precession method, with *a*- and *b*-axis zero-level precession photographs being made. The film shrinkage was calibrated by recording a silicon [111]-axis zero level precession photograph on the same film. MoK $\alpha$ -radiation was used. The accuracy achieved corresponds to probable errors of  $\pm 0.010$ ,  $\pm 0.020$ ,  $\pm 0.005$  Å, and  $\pm 0^{\circ}05'$  for *a*, *b*, *c*, and  $\beta$ , respectively. The cell dimensions are given in Table 5. Comparison with Table 4 reveals that the cell dimensions of ferrohastingsites decrease with increasing  $100\text{Mg}/\text{Mg} + \text{Fe}^{2+} + \text{Fe}^{3+} + \text{Mn}$ .

The structural formula of the amphiboles might be given as:



with two formulas to the unit cell. The amphibole asymmetric unit has 14 non-equivalent sites:

- $O_1$ -,  $O_2$ -,  $O_4$ -,  $O_5$ -,  $O_6$ - and  $O_7$ -sites are occupied by oxygen,
- $O_3$ -site by OH, F, Cl and O,
- $Si_{IV}$ - and  $Si_{IV}$ -sites are occupied by the tetrahedrally coordinated Si and Al; i.e., by the  $Z$ -type cations in the foregoing structural formula,
- $M_1$ -,  $M_2$ -, and  $M_3$ -sites are occupied by the octahedrally coordinated  $Y$ -type cations Mg, Fe<sup>2+</sup>, Mn, Al, Fe<sup>3+</sup>, Ti, etc. The  $M_1$ - and  $M_2$ -sites are four-fold, the  $M_3$ -site two-fold. Thus the sites mentioned accommodate 10 Mg, Fe, etc. cations per unit cell.
- $M_4$ -site occupied by the  $X$ -type cations is four-fold and has the coordination number six when occupied by small cations (Mg and Fe<sup>2+</sup> in cummingtonite) or coordination number eight when occupied by somewhat larger cations (Ca and/or Na in the calcium amphiboles). The site is too small, however, to accommodate the large K-cation.
- $A$ -site with the coordination number 10 can be vacant (as in tremolite) or partially or fully occupied by the  $A$ -cations K and the excess of Na which cannot be located in the  $M_4$ -site.

The  $b$ -repeat is a function of the mean sizes of the cations occupying the  $M_2$ - and  $M_4$ -sites (Colville et al., 1966). In calcium amphiboles the  $M_4$ -site is occupied by Ca and Na, which are quite equal in size. Thus, the  $b$ -axis is principally a function of the size of the cation occupying the  $M_2$ -site. Colville et al., (*op.cit.*, p. 1743) demonstrated that, if in magnesiohastingsite,  $NaCa_2Mg_4Fe^{3+}Si_6Al_2O_{22}(OH)_2$  with  $b = 17.98 \text{ \AA}$ , the small Fe<sup>3+</sup> is assumed to be located in  $M_2$ , the predicted increase in  $b$  for (ferro)hastingsite,  $NaCa_2Fe_4Fe^{3+}Si_6Al_2O_{22}(OH)_2$ , is  $0.16 \text{ \AA}$  when the  $M_2$ -site is occupied by 2 (Fe<sup>2+</sup>Fe<sup>3+</sup>) per unit cell. The observed increase is  $0.17 \text{ \AA}$  ( $b = 18.152 \text{ \AA}$ ).

We have calculated the  $b$ -repeat for the random distribution of 2Fe<sup>3+</sup> and 8Fe<sup>2+</sup> in the  $M_1$ -,  $M_2$ - and  $M_3$ -sites, and the predicted  $b$ -axis for ferrohastingsite is  $18.26 \text{ \AA}$ , i.e., the same as observed for the rapakivi ferrohastingsites. Ferrohastingsites with a similar  $b$ -axis have been previously described, e.g., from the younger granites of Nigeria by Frost (1963) and by Borley and Frost (1963) and from the nepheline-bearing rocks in the Marangudzi complex of Southern Rhodesia by Borley and Frost.

The  $c$ -repeat is related to the replacement of Si by Al (Borley and Frost, 1963, p. 662; Frost 1963, p. 382; Binns, 1965, p. 314; Colville et al., 1966, p. 1747). The change in  $c$  at a constant Si:Al ratio is related, according to Colville et al., to the occupancy of the  $M_1$ - and  $M_3$ -sites through the mechanism of kinking; the amphibole chain collapses around small ions and expands to accommodate larger ones. The  $c$ -axis shows an increase of about  $0.05 \text{ \AA}$  where Fe<sup>2+</sup> substitutes completely for Mg in the metal positions. The high  $c$ -values,  $5.34 \text{ \AA}$ , recorded for the rapakivi amphiboles thus refer to very Fe-rich varieties.

The clin amphibole value  $asin\beta$  — the unit repeat across facing double chains — is a function both of the sizes of the  $Z$ -type cations and of the sizes of the atoms in octahedral coordination between facing chains (Colville et al., 1966, p. 1748). An  $asin\beta$  of  $9.4 \text{ \AA}$  corresponds to exclusively small cations in these sites and a value of  $9.6 \text{ \AA}$  corresponds to all the large cations (*op. cit.*, p. 1752). The corresponding

figures — 9.593 Å—9.620 Å — of rapakivi ferrohastingsites are in good agreement with this.

Finally, the  $\beta$ -angle is related to the mean size of the cations in the  $M_4$ -site (Whittaker, 1960). The cummingtonite-grunerites are characterized by low values of  $\beta$  ( $\approx 102^\circ$ ), whereas the sodic and calcic amphiboles have higher values. Colville et al. (1966, p. 1737) give  $105.20^\circ \pm 0.34^\circ$  for their synthetic (ferro)hastingsite. Borley and Frost (1963) give the values of  $104.800^\circ$ — $104.991^\circ$  for the Nigerian ferrohastingsites and  $105.032^\circ$ — $105.193^\circ$  for the Marangudzi ferrohastingsites. The  $\beta$ -values ( $104^\circ 57'$ — $105^\circ 04'$ ) of rapakivi ferrohastingsites are in good agreement with these  $\beta$ -values.

### Chemical composition

Three new wet chemical analyses of the rapakivi ferrohastingsites are represented in Table 4, Nos. 2, 3 and 5. The No. 1 analysis is from the Sahama's paper (1947) and No. 4 is from Savolahti's paper (1962). All the analyses are calculated on the basis of 23 oxygen, thus eliminating the possible errors in the  $H_2O$ , F and Cl determination (*cf.*, Binns, 1965, p. 316). Chlorine has so far been yielded by only three wet chemically analysed rapakivi granite ferrohastingsites. The sum of the  $Z$ -type cations is calculated to 8 (tetrahedrally coordinated Si and Al). The remaining Al is regarded as having the coordination number 6 and thus calculated with the  $Y$ -type cations. The sum of the  $Y$ -type cations in each analysis is near its theoretical value 5. Among the  $Y$ -type cations  $Fe^{2+}$  prevails.  $Fe^{3+}$  always prevails over octahedrally coordinated Al. A comparison of the columns in Table 4 reveals that the decreasing  $Fe^{2+}$  is compensated by the increasing Mg. Mention must be made of the fact that the MgO content is highest in the ferrohastingsites from the dark-coloured rapakivi varieties (Nos. 4 and 5 in Table 4), whereas the iron-richest members of the ferrohastingsite (Nos. 1 and 2 in Table 4) occur in the most silicic types of the rapakivi. The  $X$ -type cations are mainly Ca and Na. A small part of Na and all of K constitute the  $A$ -type cations.

In composition the rapakivi hornblendes analysed correspond to ferrohastingsites, as previously pointed out by Sahama (1947) and Savolahti (1962). Compared with the theoretical ferrohastingsite formula,  $NaCa_2Fe_4^{2+}Fe^{3+}Si_6Al_2O_{22}(OH)_2$ , here is a small deficiency in the amounts of Al substitution in the tetrahedral sites, the excess of aluminium substituting the octahedrally coordinated ferric iron, part of the ferrous iron being replaced by magnesium and a small deficiency occurring in calcium and sodium.

The composition of the rapakivi ferrohastingsites is almost identical with the ferrohastingsite compositions from the younger granites of Northern Nigeria and from the nepheline-bearing rocks from the Marangudzi complex of Southern Rhodesia (Borley and Frost, 1963). The only exception is the slightly higher  $Na_2O$  contents

in the Nigerian and Rhodesian ferrohastingsites. Furthermore, the ferrohastingsites from the rapakivi resemble those from the Adirondack granitic rocks studied by Buddington and Leonard (1953). In the Precambrian of Finland hornblendes quite similar to the rapakivi ferrohastingsites have been found in the granitic rocks of the granite province in Central Finland (*cf.*, Rouhunkoski, 1959; Simonen, 1960, p. 52).

TABLE 4

Chemical analyses and atomic ratios to 23 oxygen (anhydrous basis) of the rapakivi amphiboles

	1	2	3	4	5	6	7	8
SiO <sub>2</sub> .....	38.24	39.32	40.94	39.78	41.38	40.53*	47.43*	48.90*
TiO <sub>2</sub> .....	2.00	2.05	1.60	2.32	1.94	0.23	0.16	0.01
Al <sub>2</sub> O <sub>3</sub> .....	10.17	9.91	9.46	10.14	8.96	8.81	1.75	0.24
Fe <sub>2</sub> O <sub>3</sub> .....	5.00	5.12	4.45	2.96	4.10	±30.26*	±43.95*	±46.01*
FeO .....	26.64	27.17	25.92	25.68	23.91	—	—	—
MnO .....	0.28	0.23	0.31	0.31	0.22	0.33	0.76	1.06
MgO .....	1.07	1.33	2.86	3.58	4.59	1.87	1.16	0.78
CaO .....	10.64	9.72	10.11	10.55	10.44	9.21	1.93	0.21
Na <sub>2</sub> O .....	1.50	1.43	1.65	1.62	1.74	1.55	0.24	0.12
K <sub>2</sub> O .....	1.57	1.86	1.58	1.60	1.51	1.25	0.23	0.05
H <sub>2</sub> O+ .....	1.88	1.43	1.20	1.18	1.02	—	—	—
H <sub>2</sub> O— .....	0.08	0.00	0.00	0.02	0.02	—	—	—
F .....	1.06	0.32	0.54	0.69	0.61	—	—	—
Cl .....	0.51	0.64	0.39	—	—	0.37	0.14	0.14
	100.64	100.53	101.01	100.43	100.44	—	—	—
—O .....	0.57	0.27	0.32	0.29	0.26	—	—	—
	100.07	100.26	100.69	100.14	100.18	—	—	—
Z { Si .....	6.23	6.32	6.46	6.29	6.47	6.66	7.80	8.09
Al <sup>[4]</sup> .....	1.77	1.68	1.54	1.71	1.53	1.34	.20	.00
Al <sup>[6]</sup> .....	.18	.20	.22	.17	.12	.36	.30	.06
Ti .....	.15	.25	.19	.26	.23	.03	.04	.00
Y { Fe <sup>3+</sup> .....	.61	.62	.53	.36	.48	.55]**		
Fe <sup>2+</sup> .....	3.64	3.67	3.42	3.39	3.13	3.61}	6.05	6.36
Mn .....	.04	.03	.04	.04	.03	.05	.11	.14
Mg .....	.27	.32	.67	.85	1.07	.45	.29	.19
X, A { Ca .....	1.87	1.67	1.71	1.79	1.75	1.62	.34	.04
Na .....	.47	.45	.50	.49	.53	.49	.04	.02
K .....	.33	.38	.32	.36	.30	.26	.02	.00
ΣZ .....	8.00	8.00	8.00	8.00	8.00	8.00	8.00	8.09
ΣY .....	4.89	5.09	5.07	5.07	5.06	5.05	6.79	6.75
ΣX, A .....	2.67	2.50	2.53	2.64	2.58	2.39	.40	.06
OH .....	2.04	1.53	1.27	1.24	1.07	—	—	—
F .....	.55	.16	.27	.34	.30	—	—	—
Cl .....	.14	.17	.10	—	—	.10	.02	.02
100 Mg								
Mg + Fe + Mn	5.92	6.90	14.38	18.32	22.72	9.66	4.50	2.84

1. Ferrohastingsite from pyterlite. Salmi rapakivi massif. Uuksunjoki, Salmi. Anal. O. v. Knorring (Sahama, 1947).
2. Ferrohastingsite from coarse-grained rapakivi granite. Suomenniemi rapakivi massif. No. 292/FP/54. Suomenniemi. Anal. P. Ojanperä, fluorine determined by A. Heikkinen.
3. Ferrohastingsite from dark-coloured wiborgite. Wiborg rapakivi massif. No. 785/ML/54. Tutka-asema, Valkeala. Anal. P. Ojanperä, fluorine determined by A. Heikkinen.

4. Ferrohastingsite from green marginal variety of hornblende rapakivi. Mäntyharju rapakivi massif. Nurmaa, Mäntyharju. Anal. A. Savolahti (Savolahti, 1962).
5. Ferrohastingsite from dark-coloured rapakivi granite (tirilite). Wiborg rapakivi massif. No. 35 b/AS/54. Tullisenlampi, Lemi. Anal. P. Ojanperä, fluorine determined by A. Heikkinen.
6. Ferrohastingsite rim around grunerite from the dark-coloured wiborgite. Wiborg rapakivi massif. No. 785/ML/54. Tutka-asema, Valkeala. Partial electron microprobe analysis, ferrohastingsite No. 3 (in this table) as standard. Anal. J. Siivola.
7. Grunerite from coarse-grained rapakivi granite. Suomenniemi rapakivi massif. No. 292/FP/54 (*cf.*, anal. No. 2). Suomenniemi. Partial electron microprobe analysis, ferrohastingsite No. 2 (in this table) as standard. Anal. J. Siivola.
8. Grunerite from dark-coloured wiborgite. Wiborg rapakivi massif. No. 785/ML/54. Tutka-asema, Valkeala. Partial electron microprobe analysis, ferrohastingsite No. 3 (in this table) as standard. Anal. J. Siivola.

\* corrected for absorption

‡ total iron

\*\* total iron divided between ferrous and ferric iron in the same ratio as in anal. No. 3 (ferrohastingsite from the same specimen).

TABLE 5  
Physical properties and unit cell dimensions of rapakivi amphiboles

	1	2	3	4	5	6	7
$a$ .....	1.702	1.706	1.696	1.687	1.683	—	—
$\beta$ .....	—	1.725	1.708	1.703	1.702	—	—
$\gamma$ .....	1.730	1.732	1.715	1.716	1.712	—	—
$2V\alpha$ .....	44°	24°	46°	60°	60°	—	—
$c \wedge Z$ .....	12°	18°	15°	23°	18°	11°	13°
$X$ .....	P.gr.br.	P.gr.br.	P.gr.br.	P.gr.br.	P.gr.br.	—	—
$Y$ .....	D.gr.br.	Bl.gr.	Br.gr.	Br.gr.	Br.gr.	—	—
$Z$ .....	Bl.gr.	Bl.gr.	Bl.gr.	Bl.gr.	D.gr.	—	—
$D$ (g/cc) .....	3.447	3.441	3.413	3.40	—	—	—
$a$ (Å) .....	9.937	9.962	9.942	—	9.929	9.576	—
$b$ .....	18.241	18.261	18.260	—	18.214	18.41	—
$c$ .....	5.336	5.345	5.335	—	5.321	5.344	—
$\beta$ .....	104°59'	105°04'	105°02'	—	104°57'	101°51'	—
$a \sin \beta$ .....	9.606	9.620	9.602	—	9.593	9.372	—
$V$ (Å <sup>3</sup> ) .....	935.0	939.0	935.4	—	929.7	922.0	—

P. = pale; gr. = greenish, green; D. = dark; br. = brownish, brown; Bl. = bluish.

Nos. 1—5 ferrohastingsites as in Table 4, No. 6 grunerite from the specimen 292/FP/54 and No. 7 grunerite from the specimen 785/ML/54 (*cf.*, Table 4).

### Physical properties

The optical properties, densities (by pycnometer) and cell dimensions of the rapakivi ferrohastingsites are given in Table 5 (Nos. 1—5). The optical data (except optical angle for No. 1) and densities for the ferrohastingsites Nos. 1 and 4 are taken from the papers of Sahama (1947) and Savolahti (1962). The relation between the physical properties and chemical composition of the rapakivi ferrohastingsites is presented in Fig. 2, which shows the physical properties as a function of increasing atomic ratio  $100\text{Mg}/\text{Mg} + \text{Fe}^{2+} + \text{Fe}^{3+} + \text{Mn}$  of the analysed ferrohastingsites. The data presented are in very good agreement with the diagram published by Deer

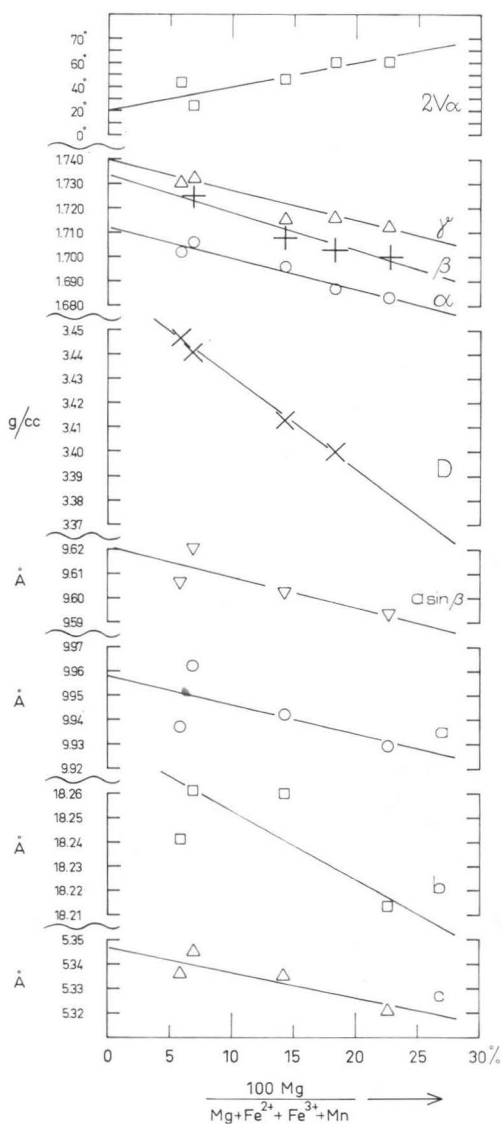


FIG. 2. Relation between the physical properties and chemical composition of the rapakivi ferro-hastingsites.  $2V\alpha$ , optical axial angle;  $a, \beta$  and  $\gamma$ , the refractive indices;  $D$ , density;  $a, b, c$  and  $a \sin \beta$ , the lattice constants.

et al. (1963, p. 298) for the relation between chemical composition and the optical data and density of the pargasite-ferrohastingsite amphiboles.



## GRUNERITE

## Occurrence

Grunerite occurs in the dark-coloured hornblende-bearing varieties of rapakivi from the different parts of the broad rapakivi area in southeastern Finland. The content of grunerite is always very low, only 0.1—0.3 vol. per cent, according to point count analyses. The Fe-Mg-silicates coexisting with grunerite are hornblende, biotite, olivine and/or iddingsite (*cf.*, Table 3). The grunerite occurs homoaxially intergrown with ferrohastingsite and it often takes the form of a rim (Fig. 3) between ferrohastingsite and iddingsite, the latter being a pseudomorph after olivine. Occasionally, the margin between ferrohastingsite and grunerite is sharp and linear, but in most cases, the grunerite occurs as irregularly shaped patches in ferrohastingsite.

Grunerite has been described from many rapakivi granites. A colourless rapakivi amphibole, later identified as grunerite, was mentioned as early as 1903 by Popov in his description of the Korsun rapakivi massif near Kiev in the Ukraine. Later, Luchitskiy (1911—1912) presented the new mineral, called kievite, from rapakivi granites in the Ukraine and Finland. The mineral kievite was the same colourless amphibole previously described by Popov. Sobolev (1947), in his monograph on the Ukrainian rapakivi granites, described grunerite, earlier termed kievite, as a



FIG. 3. Grunerite rim (*G*) between ferrohastingsite (*F*) and iddingsite (*I*). Dark-coloured wiborgite (785/ML/54). Tutka-asema, Valkeala.

mineral that occurs in many samples, though in very small quantities. The grunerite from the Ukrainian rapakivi is always accompanied by hornblende, fayalite and/or iddingsite (*op.cit.*, pp. 47—49, 110—111). The occurrence of grunerite in postorogenic granites of eastern Sayan is also reported by Sobolev (1947, p. 119).

Rouhunkoski (1959) has described a granite from Petäjävesi, Central Finland, which contains orthoclase, quartz, plagioclase, ferrohastingsite, iron-rich biotite, fayalite, orthopyroxene ( $En_{14}Fs_{86}$ ), diopsidic augite and cummingtonite, the last-mentioned mineral homoaxially intergrown with ferrohastingsite. The granitic bulk composition of the rock, which is similar to that of the dark-coloured rapakivi varieties, and its mineralogical composition characterized by extremely iron-rich silicates (fayalite, ferrohastingsite, eulite) strongly suggest in the present authors' view, that the mineral described as cummingtonite may be grunerite.

### X-ray crystallography

Grunerite (in specimen 292/FP/54), homoaxially intergrown with ferrohastingsite, was studied by X-ray methods. The Buerger precession photographs show the grunerite  $c$ -axis to be parallel to the ferrohastingsite  $c$ -axis and the grunerite  $b$ -axis parallel to the ferrohastingsite  $b$ -axis. The cell dimensions of this grunerite are given in Table 5, No. 6. A silicon single crystal has been used as a standard to eliminate the influence of film shrinkage on the  $a$ -,  $b$ - and  $c$ -values. According to the cell dimensions, when the X-ray determinative curve of Viswanathan and Ghose (1965) is applied, the content of the magnesian end member of the grunerite studied is 9 mol. per cent (100 Mg/Mg+Fe = 9 %).

### Chemical composition

Two rapakivi grunerites were investigated using an electron probe micro-analyser and the results are given in Table 4, Nos. 7 and 8.

The No. 7 grunerite in Table 4 occurs as irregular patches in ferrohastingsite (*cf.* Table 4, No. 2) and it is unevenly pigmented by some reddish yellow mineral, which may have a slight influence on the chemical composition determined. The content of the magnesian end member of the grunerite studied by micro-probe analysis is only 4—5 mol. per cent, whereas that by the cell dimensions was similar or 9 mol. per cent.

The No. 8 grunerite in Table 4 forms a narrow rim (Fig.3) between iddingsite and ferrohastingsite and its composition is near the end member composition  $Fe_7Si_8O_{22}(OH)_2$ . It should be remarked that the micro-probe analysis of the ferrohastingsite rim surrounding the grunerite rim is also presented in Table 4, No. 6; it deviates slightly from the chemical composition of the ferrohastingsite proper (Table 4, No. 3). As

for the Fe/Mg distribution between the grunerite and the ferrohastingsite, it can be stated that the grunerite is enriched in iron.

### Physical properties

Incomplete optical data (only  $c \wedge Z$ ) on the rapakivi grunerite and the cell dimensions, determined by X-ray study, are given in Table 5, Nos. 6 and 7.

## BIOTITE

### Occurrence

Biotite occurs in all varieties of the rapakivi (*cf.*, Table 3) The content of biotite in the dark-coloured hornblende-bearing varieties is usually low, only 0.3 to 4 per cent. The biotite is the principal mafic mineral of the most silicic types of the rapakivi, which generally contain 2 to 6 per cent biotite.

### X-ray crystallography

Two chemically analysed biotites (Nos. 1 and 2 in Table 6) were investigated by X-ray single crystal and powder methods. The X-ray data are given in Table 7, Nos. 1 and 2. Precession photographs show the  $1M$ -polytype of the biotites studied. The cell dimensions of biotite No. 1 in Table 7 were determined from a silicon-calibrated powder diagram using  $CuK\alpha$ -radiation with an Xe-filled proportional counter. The accuracy achieved corresponds to probable errors of 0.01, 0.02, 0.02 Å and 10' in  $a$ ,  $b$ ,  $c$  and  $\beta$ , respectively. The cell dimensions of biotite No. 2 in Table 7 were determined from precession photographs, recording on the same film a silicon [111]-axis zero level precession photograph. The probable errors are 0.005, 0.01, 0.01 Å and 15' in  $a$ ,  $b$ ,  $c$  and  $\beta$ , respectively. Twinning on [110] was observed for the biotite No. 1, Table 7.

It is now well known that the sheet dimensions of layer-lattice silicates are mainly controlled by the octahedrally coordinated cations (octahedral layer) and, especially in micas, also by the interlayer cations (Radoslovich and Norrish, 1962). Using the regression coefficients of Radoslovich (1962) for the rapakivi micas Nos. 1 and 2 in Tables 6 and 7, the calculated  $b$  is 9.318 and 9.320 Å (both  $\pm 0.03$  Å), respectively. Agreement with the observed values is excellent (*cf.*, Table 7). The observed  $b$ -values, vice versa, point to exceedingly iron-rich trioctahedral micas.

**Chemical composition**

Two new chemical analyses of rapakivi biotites are given in Table 6, Nos. 1 and 2. Rapakivi biotite analyses Nos. 3 and 4 in Table 6 are from the papers of Savolahti (1962) and Eskola (1949). All the analyses are calculated on the anhydrous basis (= atomic ratios to 22 oxygen), because chlorine is so far determined from only one rapakivi granite biotite (No. 1 in Table 6).

In composition the rapakivi granite biotites closely correspond to lepidomelane (in the sense of Foster, 1960), i.e., biotite variety characterized by a high octahedrally coordinated ferrous iron content and practically no aluminium in the octahedral voids. The amount of ferric iron in octahedral positions is significant. Magnesium is somewhat more abundantly represented than in the formulas of representative lepidomelane reported by Foster (*op.cit.*, p. 31). The four chemically analysed rapakivi granite biotites are plotted in Fig. 4 in a triangular diagram, where the fields indicating the relation between chemical composition and geologic occurrence of biotites have been drawn according to Foster (*op.cit.*, p. 33, Fig. 12). As seen, the composition points of the rapakivi granite biotites fall into a very small field. In the terminology of Foster, this field lies between three varieties, namely Fe-biotite, lepidomelane and the annite molecule ( $\text{KFe}_3[(\text{OH})_2\text{AlSi}_3\text{O}_8]$ ).

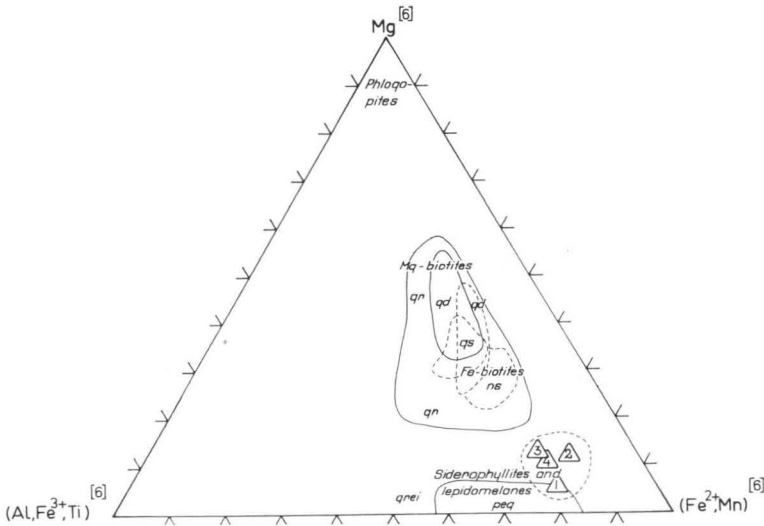


FIG. 4. Proportions of octahedrally coordinated Mg,  $\text{Fe}^{2+} + \text{Mn}$  and  $\text{Al} + \text{Fe}^{3+} + \text{Ti}$  in rapakivi biotites. Nos. 1—4 refer to Table 6. The fields showing the composition of biotites from different geological environments are presented according to Foster (1960). *gr*, granite; *gd*, granodiorite; *qd*, quartz diorite; *ns*, nepheline syenite; *gs*, gneiss and schist; *peg*, pegmatite; *grei*, greisen.

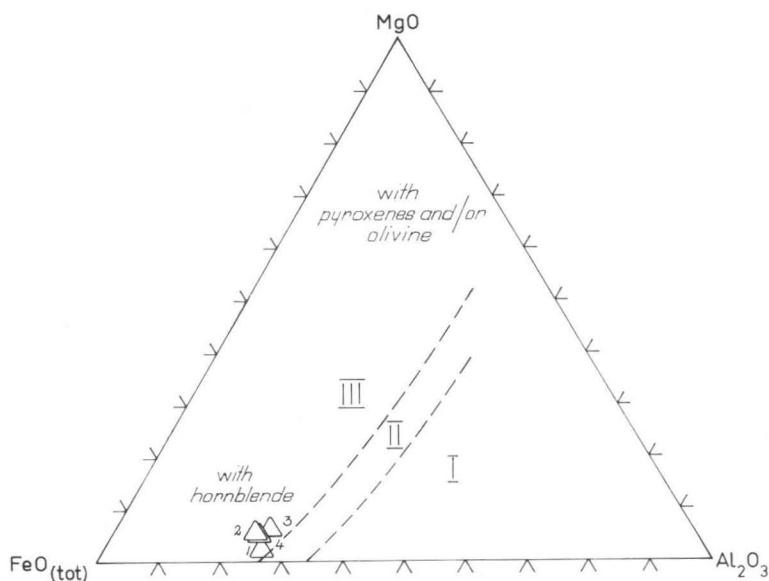


FIG. 5. Proportions of MgO,  $\text{Al}_2\text{O}_3$ , and FeO (tot) in weight percentages in the biotites. Nos. 1—4 refer to Table 6. The fields (I—III, see text) show biotite compositions for different mineral associations according to Nockolds (1947).

In Fig. 5 the chemically analyzed biotites are presented in the triangular diagram, where the three corners are represented by MgO,  $\text{Al}_2\text{O}_3$  and FeO (total iron), respectively. The three fields (I—III) show, according to Nockolds (1947), biotite compositions for different mineral associations as follows:

- I, biotites associated with muscovite, topaz, etc.,
- II, biotites unaccompanied by other mafic minerals,
- III, associated with hornblende, pyroxene and/or olivine.

The rapakivi biotites are in field III and there in the part where the biotite is mainly associated with hornblende, which is the main mafic mineral of the most common types of rapakivi.

### Physical properties

The physical data of the chemically studied rapakivi biotites are given in Table 7. The high refractive indices are similar to those of rapakivi biotites presented by Savolahti (1962). The exceedingly high iron content of the biotite causes both the high refractive indices and high specific gravity.

TABLE 6

Chemical analyses and atomic ratios to 22 oxygen (anhydrous basis) of the rapakivi biotites

	1	2	3	4
SiO <sub>2</sub> .....	34.60	34.90	35.60	34.72
TiO <sub>2</sub> .....	3.13	2.82	1.90	3.75
Al <sub>2</sub> O <sub>3</sub> .....	12.72	11.44	12.28	11.09
Fe <sub>2</sub> O <sub>3</sub> .....	4.54	3.99	4.65	6.12
FeO .....	31.18	31.44	29.47	28.96
MnO .....	0.20	0.15	0.30	0.45
MgO .....	1.31	2.80	3.14	2.44
CaO .....	0.38	0.19	1.92	0.79
Na <sub>2</sub> O .....	0.38	0.41	0.89	0.53
K <sub>2</sub> O .....	7.66	8.38	6.67	7.57
H <sub>2</sub> O+ .....	3.48	3.21	3.11	2.44
H <sub>2</sub> O- .....	0.08	0.04	0.00	0.75
F .....	0.20	0.54	0.93	0.27
Cl .....	0.62	—	—	—
P <sub>2</sub> O <sub>5</sub> .....	—	—	0.11	0.01 (V <sub>2</sub> O <sub>5</sub> )
	100.48	100.31	100.97	99.89
—O .....	0.22	0.22	0.39	0.13
	100.26	100.09	100.58	99.76
Z { Si .....	5.62	5.67	5.67	5.62
{ Al <sup>[4]</sup> .....	2.38	2.19	2.30	2.11
{ Ti <sup>[4]</sup> .....	.00	.14	.03	.27
{ Al <sup>[6]</sup> .....	.06	.00	.00	.00
{ Ti <sup>[6]</sup> .....	.38	.20	.43	.19
Y { Fe <sup>3+</sup> .....	.55	.49	.55	.74
{ Fe <sup>2+</sup> .....	4.24	4.28	3.93	3.92
{ Mn .....	.03	.02	.04	.06
{ Mg .....	.32	.68	.74	.59
{ Ca .....	.07	.03	.32	.14
X { Na .....	.12	.13	.27	.17
{ K .....	1.59	1.74	1.36	1.56
ΣZ .....	8.00	8.00	8.00	8.00
ΣY .....	5.58	5.67	5.69	5.50
ΣX .....	1.78	1.90	1.95	1.87
OH .....	3.77	3.48	3.32	1.66
F .....	.10	.27	.47	.14
Cl .....	.17	—	—	—
ΣOH,F,Cl .....	4.04	3.75	3.79	1.80
100Mg				
Mg+Fe <sup>2+</sup> +Fe <sup>3+</sup> +Mn	6.23	12.43	14.07	11.11

1. Biotite from coarse-grained rapakivi granite. Suomenniemi rapakivi massif. No. 292/FP/54. Suomenniemi. Anal. P. Ojanperä.
2. Biotite from dark-coloured wiborgite. Wiborg rapakivi massif. No. 785/ML/54. Tutka-asema, Valkeala. Anal. P. Ojanperä.
3. Biotite from green marginal variety of hornblende rapakivi. Mäntyharju rapakivi massif. Nurmaa, Mäntyharju. Anal. A. Savolahti (Savolahti, 1962).
4. Biotite from the Laitila rapakivi. Laitila rapakivi massif. Soukainen, Laitila. Anal. Elsa Ståhlberg (Eskola, 1949).

TABLE 7  
Physical properties and unit cell dimensions of rapakivi biotites

	1	2	3
$\beta = \gamma$ .....	1.705*)	1.690	1.695
$2V\alpha$ .....	very small	$\sim 16^\circ$	$20^\circ$
$Y = Z$ .....	B.br	B.br.	B.br.
$D$ (g/cc) <sup>1)</sup> .....	3.30	—	3.27
$a$ (Å) .....	5.377	5.380	—
$b$ .....	9.328	9.330	—
$c$ .....	10.235	10.277	—
$\beta$ .....	$99^\circ 56'$	$100^\circ 30'$	—
Polytype .....	1M	1M	—

B. = black, br. = brown.

\*)  $\pm 0.005$  (varies from grain to grain).

<sup>1)</sup> by pycnometer

1. Biotite. No. 292/FP/54. Suomenniemi (*cf.*, Table 6, No. 1).

2. Biotite. No. 785/ML/54. Tutka-asema, Valkeala (*cf.*, Table 6, No. 2).

3. Biotite (Savolahti, 1962). Nurmaa, Mäntyharju (*cf.*, Table 6, No. 3).

### CONCLUDING REMARKS

The Fe-Mg-silicates of rapakivi consist of olivine, monoclinic pyroxene, iddingsite, grunerite, ferrohastingsite, biotite and chlorite, whose associations in the different rapakivi varieties are given in Table 3. The occurrence, X-ray crystallography, chemical composition and physical properties of rapakivi ferrohastingsite, grunerite and biotite have been reported in this paper. Extremely iron-rich varieties of ferrohastingsite and biotite are the most common mafic minerals of rapakivi and they are commonly considered as products of the crystallization of rapakivi magma. Grunerite is exceedingly rare in magmatic rocks, but it is found in small quantities in certain dark-coloured varieties of rapakivi. The origin of rapakivi grunerite will be briefly discussed.

So far as is known, the members of the cummingtonite-grunerite series are almost exclusively restricted to metamorphic rocks, and therefore it is quite unlikely that the rapakivi grunerite would be a direct crystallization product of the silicate melt. Furthermore, it should be noted that experimentally determined phase relationships (see, e.g., Ernst, 1968, pp. 45—47) indicate that the members of the cummingtonite—grunerite series are stable only at submagmatic temperatures, whereas at temperatures above  $595^\circ\text{C}$  (Schürman, 1966) grunerite disintegrates into fayalite and quartz. The magmatic crystallization of rapakivi at a high temperature has been suggested by various authors, who have pointed out the monoclinic symmetry of rapakivi potash feldspar (orthoclase micropertthite) and the presence of idiomorphic, hexagonal bipyramidal quartz (originally high quartz). Furthermore, the feldspar-equilibrium temperature of the rapakivi is as high as  $730^\circ\text{C}$ , indicating a truly magmatic range of temperature (*cf.*, Simonen, 1961 b).

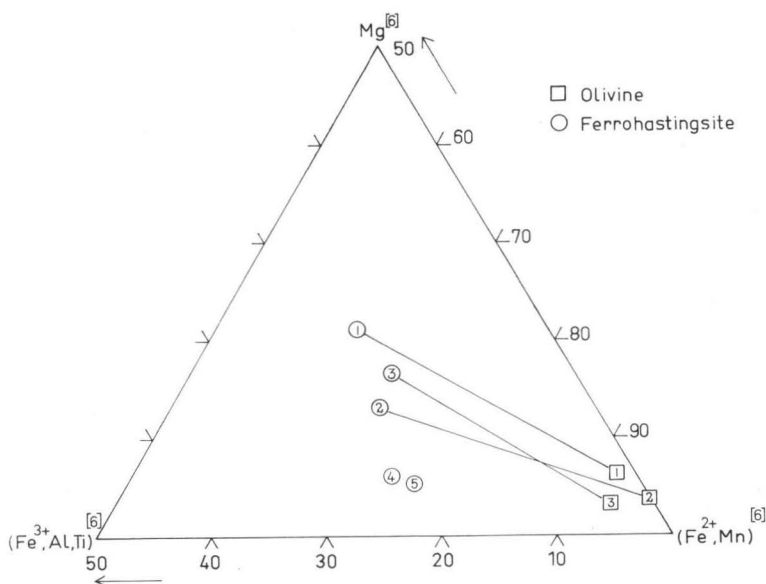


FIG. 6. Proportions of octahedrally coordinated Mg, Fe<sup>2+</sup> + Mn and Fe<sup>3+</sup> + Al + Ti in rapakivi olivines and hornblende. Coexisting mineral pairs are connected by tielines. Nos. 1—5 refer to rock specimens as follows:

1. Dark-coloured rapakivi (tirilite). Tullisenlampi, Lemi (*cf.*, Table 4, No. 5; olivine studied by Simonen, 1961 a).
2. Dark-coloured wiborgite. Tutka-asema, Valkeala (*cf.*, Table 4, No. 3; olivine studied by micro-probe analysis, *cf.*, p. 24).
3. Green marginal variety of hornblende rapakivi. Nurmaa, Mäntyharju (*cf.*, Table 4, No. 4; Savolahti, 1962).
4. Coarse-grained rapakivi granite. Suomenniemi (*cf.*, Table 4, No. 2).
5. Pyterlite. Uuksunjoki, Salmi (*cf.*, Table 4, No. 1; Sahama, 1947).

The genesis of grunerite in the Ukrainian rapakivi granites has been discussed by Sobolev (1947), who points out that the rapakivi grunerite originated at a post-magmatic stage contemporary with that of bluish-green hornblende, whereas the crystallization of grunerite took place in many different ways. In some cases, the grunerite is a result of the reaction between fayalite and quartz or a result of the oxidation process of olivine, a process that also produced magnetite. In other cases, the grunerite developed at the expense of brown hornblende so that the latter was attacked by fluorine-rich solutions and the process produced fluorite, calcite and quartz as well.

The Fe/Mg distribution of the rapakivi grunerite is quite similar to that of olivine (see Figs. 6 and 7) and, therefore, it would be reasonable to assume that the grunerite is an alteration product of olivine. Certain petrographic features found in the grunerite-bearing rapakivi studied do not, however, support this interpretation. Special attention must be drawn to the fact that the grunerite in Finnish rapakivis



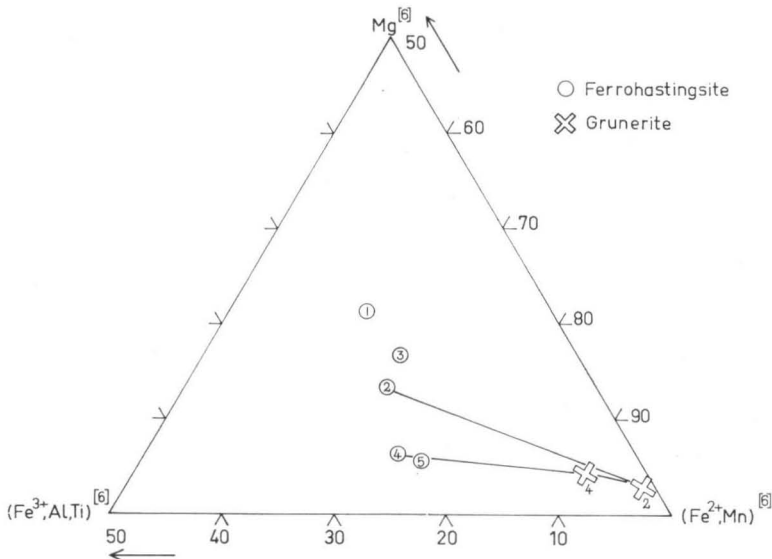


FIG. 7. Proportions of octahedrally coordinated Mg,  $\text{Fe}^{2+} + \text{Mn}$  and  $\text{Fe}^{3+}$ ,  $+\text{Al} + \text{Ti}$  in rapakivi ferrohastingsites and grunerites. Coexisting studied mineral pairs connected by tielines. Nos. 1—5 refer to specimens listed in Fig. 6.

almost always occurs homoaxially intergrown in ferrohastingsite and it is always associated with iddingsite and ferrohastingsite. The grunerite further occurs as rims between iddingsite and ferrohastingsite, and it is likewise in this case homoaxially intergrown with ferrohastingsite. The Fe/Mg distribution of the olivine and the grunerite is similar, but the grunerite differs from the ferrohastingsite in that the former is enriched in iron. Two micro-probe analyses of separate iddingsite chondres from specimen No. 785/ML/54 (see Table 4, No. 8) gave FeO 29.3, 30.2 and MgO 1.3 and 1.8 per cent, respectively. The olivine from the same specimen contains, according to micro-probe analyses, FeO 59.6 and MgO 1.3 per cent. These analytical data show that a remarkable amount of iron is expelled in the deuteric reaction producing iddingsite from olivine. It is reasonable to assume that expelled iron is now to be found in grunerite ( $\text{FeO}_{\text{tot}}$  in grunerite is about 45 per cent, whereas that in ferrohastingsite is only about 30 per cent).

Petrographic characteristics and the Fe/Mg distribution between Fe-Mg-silicates indicate that the origin of grunerite is due to deuteric or hydrothermal reactions after the magmatic crystallization of rapakivi. Reactions were induced in the ferrohastingsite by siliceous aqueous fluids at the same time as the olivine altered into iddingsite and the iron expelled during the latter process became situated in grunerite, i.e., the X-type cations Ca and Na in the  $M_4$ -site of ferrohastingsite are essentially substituted by  $\text{Fe}^{2+}$  and the Z-type Al substituted by Si, possibly producing  $\text{Ca}_3[\text{Al}(\text{OH})_6]_2$ , the latter being soluble and expelled.

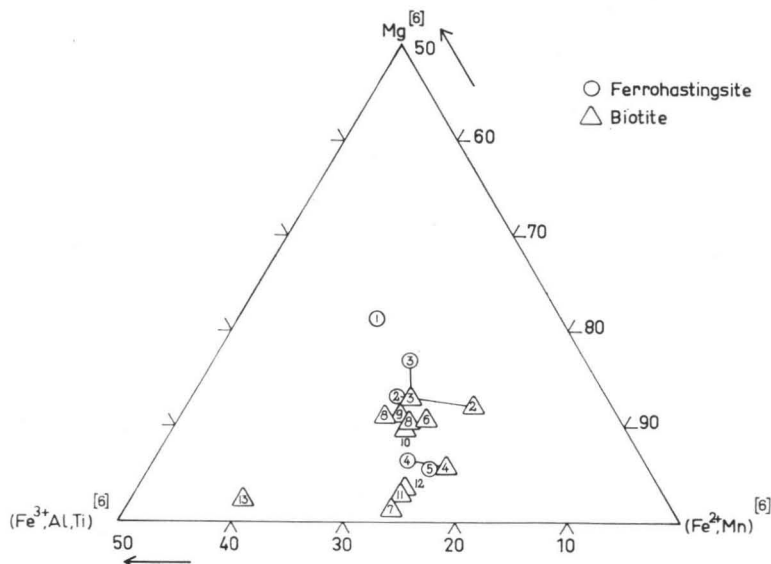
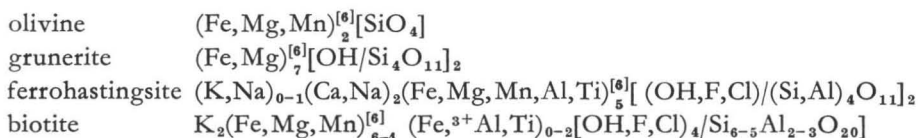


FIG. 8. Proportions of octahedrally coordinated Mg, Fe<sup>2+</sup> + Mn and Fe<sup>3+</sup> + Al + Ti in rapakivi ferrohastingsites and biotites. Coexisting studied mineral pairs connected by tielines. Nos. 1—5 refer to specimens listed in Fig. 6. Nos. 6—13 are biotites presented by Sviridenko (1967) from the rapakivi in the Salmi massif. 6—7 wiborgites; 8—9, pyterlites; 10, »coarse-ovoidal», porphyritic rapakivi; 11—12, even-grained rapakivi granites; 13, »uneven-grained» rapakivi granite.

The structure formulas of the rapakivi Fe-Mg-silicates studied chemically are as follows:



In each of the given formulas Mg, Fe<sup>2+</sup>, Mn, Fe<sup>3+</sup>, Ti and part of Al represent octahedrally coordinated cations and proportions of (Mg)<sup>[6]</sup>: (Fe<sup>2+</sup>, Mn)<sup>[6]</sup>: (Fe<sup>3+</sup>, Al, Ti)<sup>[6]</sup> are presented in Figs. 6—8. The distribution of the foregoing components among the octahedral sites of the studied minerals is regular. The simple compounds, olivine and grunerite (Figs. 6 and 7), have the same distribution, whereas the cation distribution in more complex compounds, ferrohastingsite and biotite (Fig. 8), deviates from that found in olivine and grunerite. In Figs. 6—8, the mineral pairs, which occur in the same specimen, are connected with tielines.

Three olivine-ferrohastingsite pairs presented in Fig. 6 show that ferrohastingsites coexisting with olivine are richer in MgO than those (Nos. 4—5 in Fig. 6) not coexisting with olivine. Two analysed grunerites are plotted in Fig. 7. It should be mentioned

TABLE 8  
Comparison of the compositions of coexisting rapakivi granite biotites and hornblendes \*)

	1		2		3	
	<i>Bi</i>	<i>Hbl</i>	<i>Bi</i>	<i>Hbl</i>	<i>Bi</i>	<i>Hbl</i>
Mg .....	0.68	0.67	0.32	0.32	0.74	0.85
Fe <sup>2+</sup> .....	4.28	3.42	4.24	3.67	3.93	3.39
Mg/Mg+Fe <sup>2+</sup> .....	0.137	0.196	0.070	0.087	0.158	0.200
Al <sup>[4]</sup> .....	2.19	1.54	2.38	1.68	2.30	1.74
Fe <sup>3+</sup> .....	0.49	0.53	0.55	0.62	0.55	0.36
Ti .....	0.34	0.19	0.38	0.25	0.46	0.26
Al <sup>[6]</sup> .....	0.00	0.22	0.06	0.20	0.00	0.17
Al <sup>[6]</sup> +Fe <sup>3+</sup> +Ti .....	0.83	0.94	0.99	1.07	1.01	0.79
Na .....	0.13	0.50	0.12	0.45	0.27	0.49

\*) In terms of 24 (O,OH,Cl and F) anions. *Bi* = biotite, *Hbl* = hornblende.

1. Dark-coloured wiborgite. Tutka-ascma, Valkeala. No. 785/ML/55 (*cf.*, Table 4, No. 3 and Table 6, No. 2).
2. Coarse-grained rapakivi granite. Suomenniemi. No. 292/FP/54 (*cf.*, Table 4, No. 2 and Table 6, No. 1).
3. Marginal variety of hornblende rapakivi. Nurmaa, Mäntyharju (*cf.*, Table 4, No. 4 and Table 6, No. 3).

that the iron has been determined as FeO, but probably the content of ferric iron in grunerite is quite low (*cf.*, Klein and Waldbaum, 1967, p. 380).

The cation distribution of ferrohastingsite and biotite is shown in Fig. 8, where three coexisting ferrohastingsite-biotite pairs are presented. The Mg/Fe+Mn distribution of the ferrohastingsite-biotite pairs indicate that the biotite invariably has a slightly lower Mg/Fe+Mn ratio than that of the coexisting ferrohastingsite. Biotites from the Salmi rapakivi studied recently by Sviridenko (1967) are also presented in Fig. 8 and their cation distribution is with few exceptions related to that of the biotites discussed in the paper. According to Sviridenko, a remarkable portion of the ferrous iron in rapakivi biotites has been oxidized to ferric iron, with the result that the annite component has been partly altered into oxianite ( $\text{KFe}_3^{2+}\text{AlSi}_3\text{O}_{10}(\text{OH})_2 \rightleftharpoons \text{KFe}^{2+}\text{Fe}_2^{3+}\text{AlSi}_3\text{O}_{12} + \text{H}_2$ ) and, further, the Fe<sup>3+</sup>/Fe<sup>2+</sup> ratio is quite similar in all the biotites analysed.

DeVore (1957) has shown that, with few exceptions, biotite contains more Al<sup>[4]</sup>, Fe<sup>3+</sup>, Ti and Al<sup>[6]</sup>+Fe<sup>3+</sup>+Ti (in terms of 24(O, OH and F) anions) than does the coexisting hornblende, the Mg/Mg+Fe<sup>2+</sup> ratio of the biotite is lower than that of the coexisting hornblende and, furthermore, the hornblende contains more Al<sup>[6]</sup> and Na than does the coexisting biotite. The data observed in the coexisting rapakivi hornblende and biotite (Table 8) are in quite good agreement with the conclusions of DeVore just cited. The only exception is that the Fe<sup>3+</sup> content in some rapakivi hornblendes is slightly higher than that of coexisting biotites. As a result, furthermore, the content of Al<sup>[6]</sup>+Fe<sup>3+</sup>+Ti in hornblendes may be slightly higher than that of the coexisting rapakivi biotites. The over-all agreement with DeVore's results points to the equilibrium of these two phases.

## REFERENCES

- BINNS, R. A. (1965) The mineralogy of metamorphosed basic rocks from the Willyama Complex, Broken Hill district, New South Wales. Part I. Hornblendes. *Mineral. Mag.*, Vol. 35, No. 270, 306—326.
- BORLEY, G., and FROST, M. T. (1963) Some observations on igneous ferrohastingsites. *Mineral. Mag.*, Vol. 33, No. 263, 646—662.
- BUDDINGTON, A. F., and LEONARD, B. F. (1953) Chemical petrology and mineralogy of hornblendes in northwest Adirondack granitic rocks. *Amer. Mineral.*, Vol. 38, Nos. 11 — 12, 891—902.
- COLVILLE, PATRICIA A., ERNST, W. G., and GILBERT, M. C. (1966). Relationships between cell parameters and chemical compositions of monoclinic amphiboles. *Amer. Mineral.*, Vol. 51, Nos. 11—12, 1727—1754.
- DEER, W. A., HOWIE, R. A., and ZUSSMAN, J. (1963) *Rock-forming minerals*, Vol. 2, Chain silicates, Longmans.
- DEVORE, GEORGE W. (1957) The association of strongly polarizing cations as a major influence in element distribution, mineral composition, and crystal growth. *Jour. Geology*, Vol. 65, No. 2, 178—195.
- ERNST, W. G. (1968) *Amphiboles. Minerals, Rocks and Inorganic Materials. Monograph Series of Theoretical and Experimental Studies. Vol. 1.* Springer-Verlag New York Inc.
- ESKOLA, PENTTI (1949) The mica of the moro. *Bull. Comm. géol. Finlande* 144, 113—116.
- FOSTER, MARGARET D. (1960) Interpretation of the composition of trioctahedral micas. U.S. Geological Survey, Professional Paper 354-B, 11—49.
- FROST, M. T. (1963) Amphiboles from the younger granites of Nigeria. Part II. X-ray data. *Mineral. Mag.*, Vol. 33, No. 260, 377—384.
- HACKMAN, VICTOR (1934) Das Rapakivirandgebiet der Gegend von Lappeenranta (Willmanstrand). *Bull. Comm. géol. Finlande* 106, 1—82.
- KLEIN, C., and WALDBAUM, DAVID R. (1967) X-ray crystallographic properties of the cummintonite-grunerite series. *Jour. Geology*, Vol. 75, No. 4, 379—392.
- LUCHITSKIY, V. I. (1911—1912).
- ЛУЧИЦКИЙ, В. И. (1911—1912) Материалы по петрографии юга России. Часть 1. Рапакиви Киевской губ. и породы, его сопровождающие. *Извест. Варшавск. политехн. ин-та*, вып. 2, 1911 г., стр. 1—96, и вып. 1, 1912 г., стр. 96—332.
- NOCKOLDS, S. R. (1947) The relation between chemical composition and paragenesis in the biotite micas of igneous rocks. *Amer. Journ. Sci.*, Vol. 245, 401—420.
- ПОПОВ, БОРИС (1903).
- Попов, Борис (1903) О южно-русском рапакиви. *Труды С.-Петербур. общ. естествоисп.*, т. 31, вып. 5, стр. 77—266.
- RADOSLOVICH, E. W. (1962) The cell dimensions and symmetry of layer-lattice silicates. II. Regression relations. *Amer. Mineral.*, Vol. 47, Nos. 5—6, 617—636.
- RADOSLOVICH, E. W., and NORRISH, K. (1962) The cell dimensions and symmetry of layer-lattice silicates. I. Some structural considerations. *Amer. Mineral.*, Vol. 47, Nos. 5—6, 599—616.

- ROUHUNKOSKI, PENTTI (1959) Petäjäveden alueen geologiaa. Unpublished thesis. Archives of the Dept. of Geology and Mineralogy, University of Helsinki.
- SAHAMA, TH. G. (1945) On the geochemistry of the east Fennoscandian rapakivi granites. Bull. Comm. géol. Finlande 136, 15—67.
- »— (1947) Rapakivi amphibole from Uuksunjoki, Salmi Area. Bull. Comm. géol. Finlande 140, 159—162.
- SAVOLAHNTI, ANTTI (1962) The rapakivi problem and the rules of idiomorphism in minerals. Bull. Comm. géol. Finlande 204, 33—111.
- SCHÜRMAN, K. (1966) Synthesis and stability field of cummingtonite. Cambridge, IMA-abstract.
- SIMONEN, ANTTI (1960) Plutonic rocks of the Svecofennides in Finland. Bull. Comm. géol. Finlande 189, 1—101.
- »— (1961 a) Olivine from rapakivi. Bull. Comm. géol. Finlande 196, 371—376.
- »— (1961 b) Feldspar-equilibrium temperature of some Finnish rocks. Bull. Comm. géol. Finlande 196, 367—370.
- SOBOLEV, VLADIMIR (1947)
- Соболев, Владимир (1947) Петрология восточной части сложного Коростеньского плутона. Уч. зап. Львовск. унив., т. 6, сер. геолог., вып. 5, стр. 1—137.
- SVIRIDENKO, L. P. (1967)
- Свириденко, Л. П. (1967) Петрология гранитов рапакиви Салминского массива. Известия Академии Наук СССР, Серия геологическая, вып. 2, стр. 40—55.
- VISWANATHAN, K., and GHOSE, SUBRATA (1965) The effect of  $Mg^{2+}$ — $Fe^{2+}$  substitution on the cell dimensions of cummingtonites. Amer. Mineral., Vol. 50, Nos. 7—8, 1 106—1 112.
- WHITTAKER, E. J. W. (1960) The crystal chemistry of amphiboles. Acta Cryst., Vol. 13, 291—298.

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