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CHEMICAL PETROLOGY OF SOME GRENVILLE
SCHISTS NEAR FERNLEIGH, ONTARIO.

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CHEMICAL PETROLOGY OF SOME GRENVILLE SCHISTS NEAR
FERNLEIGH, ONTARIO

by

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A thesis submitted to the Faculty of
Graduate Studies in partial fulfilment
of the requirements for the degree of
Master of Science

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ABSTRACT

Regionally metamorphosed pelitic schists, part of a Grenville metasedimentary suite at Fernleigh, Ontario, were studied petrographically and chemically. Those schists containing staurolite were studied in detail.

Eleven schists, eight biotites, seven staurolites and seven garnets were analysed chemically, largely by colorimetric and titrimetric methods.

Phase rule considerations and distribution relations indicate that the rocks attained equilibrium locally during metamorphism. Partial oxygen pressure varied among samples and may have been controlled by the original composition of the rock. Silicate mineral assemblages are affected by $\sum R_2O_3 : \sum RO$ ratio and by the concentration of Mn. Opaque mineral assemblages are mainly affected by partial oxygen pressure, the activities of S and TiO_2 playing a minor part.

Fe:Mg ratios of ferromagnesian minerals are primarily dependent on partial oxygen pressure, which determines the ferrous-ferric ratio, or the availability of ferrous iron in the rock.

Distribution of Fe and Mg between staurolite and biotite is non-ideal. Limited substitution of Mg for Fe (15-35 mole % Mg) is indicated in the staurolite structure.

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Richard Levick aided considerably in the analytical work; Walter Yzerdraat prepared the polished thin sections and assisted with mineral separations and Leo Fletcher ground the samples and performed initial mineral separations. The photomicrographs were taken by F. J. Cooke.

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CHEMICAL PETROLOGY OF SOME GRENVILLE SCHISTS NEAR FERNLEIGH, ONTARIO.

INTRODUCTION

Grenville metasedimentary rocks near Fernleigh, Ontario, are mainly pelitic schists, graphitic schists, marble and conglomerate, in the almandine amphibolite facies of regional metamorphism. The object of the present study was to obtain some knowledge of the chemical petrology of the schists, with particular emphasis on those containing staurolite.

Location

The area of study is 70 miles southwest of Ottawa, Ontario (Fig. 1), in Barrie and Clarendon townships. The area is accessible from highways 506 and 509, and from power line service roads.

Previous Investigations

Miller and Knight (1913) mapped a small area including the Ompah syncline (Fig. 2). Township mapping for the Ontario Department of Mines has been carried out by Meen and Harding (ODM Map 51d, 1944) and Smith and Peach (ODM Map 1956-4, 1958). Hewitt compiled the geology of the area (ODM Map 2053, 1964). The pelitic schist belt and adjacent units (Fig. 2) have been mapped from the west end near Myer Cave, east to the nose of the Ompah syncline, by J. M. Moore (unpublished) at a scale of one-quarter mile to the inch.

Figure 1. Location map of area studied.

GENERAL GEOLOGY

This summary of the geology of the area was obtained from a considerably more detailed account by J. M. Moore (unpublished).

The area studies lies in the 'Ottawa Circle' of the Grenville tectonic province (Wynne-Edwards, 1964), that is, in that part of the region characterized by a meta-sedimentary suite including marble. It lies on the boundary between the Kaladar-Dalhousie trough to the south and the Madawaska Highlands to the north (Hewitt, 1956).

Geology of the area is shown in Fig. 2. Lithologic units are persistent for tens of miles along strike. A belt composed dominantly of marble, paragneiss, and amphibolite is flanked on the north and south by granitic bodies. Within this belt, a band up to three-quarters of a mile wide and at least twenty-five miles long consists of interlayered mica schist, marble, and graphitic schist. Plutonic rocks in the area are grossly concordant, with foliation and lineation concordant with those of the sedimentary rocks.

Although there are variations in order and thickness of rock units in the pelitic schist belt, the stratigraphy near Fernleigh is typical. From north to south, the lithology and outcrop width (in feet) are:

Figure 2. Geological map showing localities of specimens studied. Geology slightly modified by J. M. Moore from Meen (1944) and Smith (1958).

Grey, graphite-free, porphyroblastic mica schist	(400)
Grey, dolomitic marble	(0-100)
Black, graphitic mica schist	(300)
Grey, banded, calcitic marble	(600)
Black, thinly banded, graphitic, calcareous mica schist	(2600)

The graphite-free schist, the principal topic of this thesis, is the northernmost unit everywhere along the belt, and varies from 200 to 400 feet in thickness. The thickness and order of the marble and graphitic schist layers are variable; near Myer Cave these rocks are interlayered with subordinate graphite-free schist and marble conglomerate. All analysed samples were collected from the main graphite-free schist horizon.

The schists increase in metamorphic grade from south-west to north-east, from a staurolite-kyanite zone to a sillimanite zone. Chlorite, in the form of discrete crystals, is mainly restricted to the area south-west of Fernleigh, in which it may occur with any of the other minerals of the schist. Sillimanite is seen north of Ardoch and to the north-east; it is not known to occur to the south-west. The isograds marking the eastern limit of staurolite and kyanite, and the western limit of sillimanite appear to coincide and lie just west of Ardoch. The mineral assemblages are those of the almandine-amphibolite facies (Turner & Verhoogen, 1960). In the

western part of the belt, they lie in the staurolite-almandine subfacies. East of Ardoch the sillimanite-almandine-muscovite subfacies is represented.

PETROLOGY OF THE SCHISTS

Sampling

The rocks studied are from west of the sillimanite isograd; most of the textural and mineralogical variants of these rocks are represented. Eleven graphite-free schists, and minerals from eight, were analysed.

Samples are numbered consecutively from south-west to north-east. Those collected at the same locality are distinguished by letter suffixes, for example 7A, 7B.

Field numbers and precise sample localities are listed in Appendix A.

Petrography

Petrographic methods

Extensive use was made of polished thin sections in order to identify the opaque minerals and to determine their relationship to the silicate minerals. Modal analyses were carried out with a Swift automatic point counter. The opaque mineral grains were counted by traversing a polished thin section under a reflecting microscope until approximately 100 grains were recorded. The ratio of ilmenite to hematite in intergrowths was estimated visually.

The study of textures was facilitated by using a microfilm projector having a magnification of 23.3X and a screen size of 30 x 30 cm. A portion of the thin section 13 mm square could be viewed. The average size

of some of the larger porphyroblasts was measured with a scale.

Description

(a) Graphite-free schists. (Plates 1 - 5)

Those studied consist of the mineral assemblage:

Quartz-muscovite-biotite-garnet-staurolite-ilmenite+ hematite intergrowth, with or without kyanite, plagioclase, magnetite, rutile, pyrite.

The rocks are porphyroblastic with a foliated lepidoblastic groundmass of muscovite, elongate quartz, and occasionally biotite. The porphyroblasts commonly reach a size of 5 mm; in some samples they are as large as 6 cm. The rocks containing kyanite are texturally indistinguishable from those in which kyanite is absent. The grain size of the minerals is shown in Table 1.

Quartz is anhedral, commonly elongated and polygonal. The grains vary from 0.1-0.3 mm in diameter, the larger grains occurring in rocks containing relatively large garnet and staurolite. In the groundmass quartz typically shows undulatory extinction but inclusions in the porphyroblasts generally show uniform extinction. The grains occurring as inclusions are smaller than those in the associated groundmass.

Muscovite occurs as thin plates 0.03-1.3 mm long, its size being proportional to that of the associated

Table 1

Mean grain size of minerals in schists (mm)

Graphite-free schists

Sample number	Qtz	Mus	Bio	Gar	Str	Pl	Ky	Im	Chl
1	0.2	0.03	0.2	-	0.5	0.8	-	0.01	-
2A	0.1	0.6	0.4	3.2	0.8	-	-	0.3	-
2B	0.03	0.1	0.2	-	-	0.5	3.0	0.05	0.1
6	0.1	0.1	-	-	-	-	3.0	0.1	0.15
7A	0.2	0.3	0.8	1.5	0.8	0.7	2.0	0.2	-
7B	0.2	0.2	2.0	0.8	1.7	0.8	2.0	0.1	-
7C	0.2	0.3	5.0	0.8	1.3	1.6	-	0.2	-
7D	0.3	0.5	1.0	1.5	0.8	0.8	-	0.1	-
8	0.2	0.7	1.3	1.7	6.4	1.8	-	0.1	-
9A	0.3	0.5	0.8	5.4	3.2	-	-	0.05	-
9B	0.3	0.7	2.0	3.0	2.0	0.9	-	0.1	-
10	0.2	0.3	0.4	1.5	0.4	0.3	1.0	0.1	-

Graphitic schists

Sample number	Qtz +Pl	Mus	Bio	Gar	Cc	Im
3A	0.05	0.05	0.2	1.5	-	0.03
3B	0.05	0.05	0.3	1.5	-	0.03
3C	0.05	0.05	0.2	1.0	0.05	0.6
4	0.05	0.05	0.8	2.0	-	0.03
5	0.05	0.05	0.1	0.7	0.08	0.03

Qtz - quartz in groundmass, diameter.
 Mus - muscovite, long dimension.
 Bio - biotite, long dimension.
 Gar - garnet, diameter.
 Str - staurolite, long dimension.
 Pl - plagioclase, diameter.
 Ky - kyanite, long dimension.
 Im - ilmenite or hematite, long dimension.
 Chl - chlorite, long dimension.
 Cc - calcite, diameter.

quartz. Where present in sufficient quantity it gives rise to a very distinct foliation which wraps around most of the porphyroblastic minerals.

Plagioclase occurs as subhedral to anhedral porphyroblasts ranging from 0.3-1.6 mm mean diameter. Most of the grains contain numerous quartz inclusions and show some alteration to an unknown yellow mineral, particularly along grain boundaries and cleavages. In most grains twinning is either indistinct or absent. An exception is sample 9B which contains Albite and Pericline twins. Optical determinations on three grains in this sample giving centred bisectrix figures indicate a composition of approximately An₂₅.

Garnet occurs as 0.8-2.0 mm diameter euhedral porphyroblasts containing many minute inclusions, usually confined to the centre of the crystal, or as 2-5 mm diameter subhedral to anhedral porphyroblasts containing 20-50% anhedral quartz and opaque inclusions. The groundmass where present is always deflected around garnets.

Staurolite occurs in two habits. It forms euhedral porphyroblasts ranging in size from 0.8-1.7 mm containing numerous small inclusions, usually confined to the centre of the crystal, or anhedral porphyroblasts 1.7-6.4 mm long containing up to 50% anhedral quartz and numerous opaque inclusions. Some thin sections contain both types.

The groundmass foliation may be pushed aside or intersected. In sample 10 the staurolite occurs as small anhedral, ragged crystals. Cruciform twins are common. The pleochroic formula is:

Z = golden yellow

X = Y = pale yellow to almost colourless

Biotite occurs in flakes 0.2-5 mm diameter, which in some cases show oriented opaque inclusions parallel to those in the groundmass. Many grains also contain a large number of small unidentified inclusions giving rise to large pleochroic haloes. Minor alteration to chlorite is seen in some thin sections. Porphyroblastic biotite inclined to the foliation is common and many grains deflect the foliation. Biotite in all slides has the pleochroic formula:

X = pale yellow

Z = Y = dark greenish brown

Kyanite where present occurs as prisms 0.3-1.3 mm wide and 1.5-4 mm long. It is euhedral to subhedral with few inclusions or anhedral and extremely poikiloblastic with quartz as in sample 7B.

Opaque minerals are principally ilmenite or lamellar ilmenite-hematite intergrowths in various proportions, considered to be the result of exsolution. In addition most rocks contain either magnetite or rutile and

occasionally pyrite. The ilmenite-hematite intergrowths occur as laths 0.1-0.4 mm long whereas the magnetite typically forms euhedral to anhedral equant grains 0.2-0.4 mm in diameter. Rutile prisms, generally subhedral to anhedral, are 0.1-0.2 mm long, and pyrite forms rounded blebs of highly variable size, even in a single thin section. There is no obvious concentration of any of the opaque minerals in or around any of the silicate minerals.

(b) Graphitic garnet-biotite schists. (Plate 6)

Those studied consist of the mineral assemblage:

Quartz-muscovite-biotite-garnet-graphite-ilmenite-pyrrhotite, with or without plagioclase, calcite, rutile.

In these rocks garnet is typically the only porphyroblastic mineral, and it is generally more nearly euhedral and less poikiloblastic than the garnets in the graphite-free schists. The garnets, however, are of a size range similar to those in the graphite-free rocks.

The groundmass consists of very fine quartz and muscovite, 0.05 mm, with biotite grains varying from 0.1-0.8 mm long. The sizes of the minerals in several typical graphitic schists are listed in Table 1.

The pleochroic formula of the biotite in the graphitic rocks is:

X = pale brown to yellow

Z = Y = orange to reddish brown

A.

1 mm

B.

Plate 1. Garnet-staurolite-biotite schist (7D). Euhedral garnet (G) and staurolite (S), each having numerous fine inclusions concentrated mainly in the centre of the porphyroblasts. A: plane polarized light; B: crossed polarizers.

A.

1 mm

B.

Plate 2. Kyanite-garnet-staurolite-biotite schist (7A). Kyanite grains (K) exhibit 001 parting. Staurolite is finer than kyanite and not readily recognizable from the photographs. A: plane polarized light; B: crossed polarizers.

A.

1 mm
└───┘

B.

Plate 3. Kyanite-chlorite schist (6). Sheaves of slender kyanite prisms in foliated matrix of quartz, muscovite and chlorite. A: plane polarized light; B: crossed polarizers.

A.

1 mm

B.

Plate 4. Garnet-staurolite-biotite schist (7B).
 A. Garnet, staurolite, plagioclase and quartz. Crossed polarizers.
 B. Poikiloblastic staurolite, biotite (B) and rutile. Plane polarized light.

A.

1 mm

B.

Plate 5. Kyanite-garnet-staurolite-biotite schist (10).
A. Foliated biotite and muscovite enclosing staurolite.
B. Euhedral kyanite with biotite, muscovite and quartz.
Plane polarized light.

A.

1 mm

B.

Plate 6. Graphitic garnet-biotite schist (4). The graphitic layers are deflected by garnet and pass through biotite. A: plane polarized light; B: crossed polarizers.

The garnets typically are free of graphite and deflect the matrix foliation. Half of the samples contain calcite. Twinned plagioclase is not common but where seen it is similar in size to the quartz.

The opaque minerals are ilmenite with no hematite intergrowths, pyrrhotite and usually pyrite. A few grains of chalcopyrite were seen.

Methods of representation

In the following pages modal and chemical analyses are presented. The main object and difficulty is to reduce these data to a form whereby systematic variations may be detected. The graphical methods used in this study are described below.

Three component systems

The trilinear representation of three homogeneous coordinates in either an equilateral or right isosceles triangle is possible. In most cases the latter has been preferred, for greater ease in plotting and interpretation.

Four and five component systems

(a) Successive projections.

Thompson (1957) advocated a method of representing 4 or 5 (or more) component systems by a method of successive projections in order to reduce the plot to 3 components. This can only be done if there is for each

projection one phase of essentially fixed composition common to all assemblages present. Where possible, this technique is preferable to the method described below (b), because it is readily interpretable in terms of the phase rule.

(b) Vectorial representation.

Korzhinskii (1959, p.52-60) described a method of representing multicomponent systems which has never gained popularity in the western world, despite several obvious advantages.

Composition, represented as a homogeneous bar graph, can be plotted as a vector on a right isosceles triangle, each side of the triangle being the total length of the bar graph. Using this system, a rock or mineral consisting of three components is represented by a point, four components by a vector parallel to one side of the triangle, and five components by an inclined vector. The relationship of a bar graph composed of five components a, b, c, d and e, to the vector A-A', is shown in Figure 3. A more detailed derivation of the vector diagram is given in Appendix B.

The mutual relationships of a series of samples can thus be shown diagrammatically, and the detection of any systematic variation is considerably facilitated. The relationship of ratios of any two components represented on such a diagram can be shown as a series of lines

Figure 3. Relationship between five component bar graph and five component vector diagram.

emanating from a point, the slope of each line representing the ratio. These auxiliary plots may be called ray diagrams.

Modal analysis

Complete modal analyses of the schists studied are shown in Table 2. The major minerals: muscovite, biotite, garnet, staurolite and opaque minerals, but excepting quartz, were summed, recalculated to 100% and plotted on a vector diagram in Fig. 4. From this vector plot it can be seen that the sum of biotite and muscovite remains relatively constant (a reflection of relatively constant K_2O content), whereas the biotite-muscovite ratio varies considerably. This variation is not obviously accompanied by changes in proportions of any of the other minerals, and does not exhibit any consistent areal relationship, but as the proportion of biotite to muscovite increases the grain size of most of the minerals in the rock increases. This relation is illustrated in Fig. 5 where the biotite-muscovite ratio is plotted against the size of the garnet grains, the latter being chosen because of the relatively high precision of measurement. A possible explanation is illustrated by Fig. 6, where the MnO content of garnet is plotted against its diameter; the inverse correlation obtained agrees with that observed by Chinner (1960).

Table 2
Chemical and modal analyses of schists

	1	2A	2B	6	7A	7B	7C	7D	8	9A	9B	10
SiO ₂ *	63.6	59.4	62.5	60.4	47.4	65.7	62.1	61.2	57.6		54.2	66.3
Al ₂ O ₃ *	17.9	18.7	18.2	19.7	30.4	19.4	18.0	18.7	22.5		23.7	16.4
Fe ₂ O ₃	3.6	3.5	10.5	9.7	7.0	2.8	7.7	4.3	5.2		5.3	2.4
FeO	2.6	10.7	0.0	1.8	1.2	1.4	2.8	4.1	5.1		6.2	4.2
MgO	1.4	2.3	0.08	0.24	0.13	2.1	2.0	2.7	3.5		3.5	2.8
CaO	1.9	0.52	1.1	0.72	1.5	1.2	0.38	1.4	0.50		0.76	0.73
Na ₂ O	1.3	0.12	1.1	1.1	1.3	1.2	0.52	0.33	0.26		0.52	0.71
K ₂ O	3.1	2.5	3.2	3.0	4.1	2.3	2.6	2.4	2.0		2.3	2.9
TiO ₂	1.4	1.5	1.6	1.6	2.7	1.5	1.8	1.3	1.4		1.7	1.4
MnO	0.06	0.03	0.01	0.01	0.24	0.20	0.18	0.27	0.19		0.27	0.12
P ₂ O ₅	0.20	0.12	0.11	0.12	0.19	0.06	0.08	0.55	0.30		0.07	0.14
LOI *	2.3	1.5	2.3	2.6	3.8	2.0	1.9	1.9	1.6		1.8	1.7
Total	99.4	100.9	100.7	101.0	100.0	99.9	100.1	99.2	100.2		100.3	99.8
$\frac{2Fe_2O_3 \times 100^1}{2Fe_2O_3 + FeO}$	55.5	22.7	100	82.9	84.0	64.3	71.2	48.5	47.8		43.5	33.9

Analyst A.W.H., * R. Levick
1 Oxidation ratio (Chinner, 1960).

Quartz	32.9	25.6	20.0	26.6	13.8	41.7	31.4	38.4	47.8	30.7	23.1	43.5
Muscovite	38.0	3.6	59.0#	24.5	41.9	16.6	28.3	13.2	7.5	0.1	4.0	13.3
Biotite	6.5	34.1	0.8	-	2.5	6.6	11.2	16.6	16.7	11.4	26.7	28.5
Garnet	-	7.1	-	-	5.3	0.4	0.5	6.2	0.3	26.8	8.1	1.4
Staurolite	1.3	24.3	-	-	4.3	14.9	13.2	20.6	22.4	24.3	24.0	1.4
Plagioclase	17.5	-	4.0	-	3.9	17.0	2.6	1.3	1.0	-	8.9	1.9
Kyanite	-	-	4.7	22.6	19.2	0.4	-	-	-	-	-	6.3
Opaque	3.8	5.3	11.5	12.5	9.1	2.4	12.8	3.7	4.1	6.7	5.2	3.6
Tourmaline	+	+	+	+	+	+	+	-	+	-	-	+
Apatite	-	-	-	-	+	+	+	+	+	-	-	+
Chlorite	-	-	-	14.0	-	-	-	-	-	-	-	-
Points counted	2736	3894	1316	3038	2829	4788	2803	1594	5690	2024	1197	5396
Number of slides	2	2	1	3	2	4	2	1	4	1	1	4

Chlorite and muscovite

Magnetite	43	-	-	22	-	-	+	8	8	-	-	14
Ilmenite	56.5	100	2	5	6.5	3.5	10	87.5	69	100	78	81.5
Hematite	0.5	-	98	73	85.5	66.5	90	4.5	23	-	1	4.5
Rutile	-	-	-	-	8	30	-	-	-	-	21	-
Pyrite	-	+	-	-	-	-	-	+	+	+	+	-
Points counted	102	-	-	148	83	118	-	173	91	-	94	126

Figure 4. Modal variations of staurolite schists illustrated by means of a vector diagram.

Figure 5. Correlation between diameter of garnet and biotite: muscovite ratios in staurolite schists.

Figure 6. Correlation between diameter of garnet and its Mn content.

Figs. 5 & 6 indicate that schists with a high muscovite to biotite ratio contain small garnets rich in manganese.

The proportions of garnet, biotite and staurolite are shown in Fig. 7, from which it can be seen that kyanite content is apparently independent of the ratios of the other minerals. Several of the rocks contain little garnet and one, sample 10, contains very little garnet and staurolite.

The opaque mineral modes were plotted as volume percent on a triangular diagram (Fig. 8). It can be seen that the $\text{FeO}:\text{Fe}_2\text{O}_3$ ratio varies widely but that samples are absent from the centre part of the diagram. The presence of exsolution intergrowths of hematite and ilmenite demonstrates the existence of a miscibility gap. The absence of two coexisting hematite-ilmenite intergrowths with differing proportions of the two phases suggests that bulk compositions lying within this gap were not observed.

Bulk chemical composition

Analytical methods

Chemical analyses were performed by the writer, largely by methods he has used for a number of years. These are mostly based on the methods of Shapiro and Brannock (1956) and Riley (1958). The preparation and analysis of the samples is discussed in Appendices C and D.

Chemical analyses

The bulk chemical analyses of the schists are listed in Table 2.

The facies principle, a basic axiom of metamorphic petrology introduced by Eskola (1914), is that for a given temperature and pressure the mineral assemblage is a function of the chemical composition of the rock. Traditionally this relation is shown graphically by means of ACF, AKF, and AFM diagrams.

The chemical composition of a rock is usually expressed in terms of approximately 12 constituents. To show this multicomponent system graphically in two dimensions it is usually simplified to no more than 3 components. Accordingly the silicate and opaque minerals are generally shown on separate diagrams. A complication is the presence of FeO as a major component in each group. It is not usually considered practical to determine chemically the percent FeO in the opaque minerals so an estimate is usually made on the basis of a modal analysis of the rock (Eskola, 1914). Unless the proportion of opaque minerals is small this procedure introduces a potentially large error.

Considering the non-opaque group, it is possible to reduce the number of components by excluding those occurring in only one mineral and virtually absent from the other minerals. In this study P_2O_5 , TiO_2 , Na_2O and CaO

Figure 7. Modal variations in ferromagnesian minerals.

Figure 8. Compositional variation of opaque mineral assemblages in staurolite schists, plotted as volume percent.

occur only in apatite, rutile and plagioclase respectively. Similarly trace elements, which substitute for major elements may be excluded because variations in trace element abundance normally do not affect the mineralogy of the rock.

In the rocks under consideration MnO does not form a separate mineral and in all cases except garnet occurs in minor amounts and is provisionally excluded. Silica may also be neglected, despite the fact that it enters into other minerals in large amounts, because it is present in excess, as quartz, in all rocks considered. Variations in the amount of quartz have no effect on the mineralogy of the rock (Thompson, 1957).

Fe_2O_3 occurs in small amounts in the silicate minerals but is present in major amounts in the opaque group, in hematite and magnetite. As the opaque minerals in the rocks studied are relatively abundant and their proportions are highly variable an accurate estimate of the FeO in them could not be obtained. Consequently the usual methods of illustrating the bulk composition of the silicates could not be used. The fact that the plot of bulk composition is not corrected for opaque minerals is in one way an advantage in that the plot illustrates pre-metamorphic compositions (in the absence of any evidence of metasomatism). The oxidation state of iron may, of

course, have been changed by reduction or oxidation during or after metamorphism.

The components chosen to represent the chemical compositions of the rocks are FeO , Fe_2O_3 , Al_2O_3 , MgO and K_2O , the molar proportions of which, recalculated to 100%, are shown on the vector diagram in Fig. 9. Ray diagrams showing $\text{FeO}:\text{Fe}_2\text{O}_3$ and $\text{FeO}:\text{MgO}$ ratios are shown in Fig. 10. As a further means of comparing the samples the components MgO , K_2O and Al_2O_3 were summed, recalculated to 100% and plotted on a triangle (Fig. 11). FeO and Fe_2O_3 were multiplied by the same factor and shown as auxiliary vectors (Korzhinskii, 1959, p. 54). The vector diagram and ray plots show that the rocks analysed have a very wide range of $\text{FeO}:\text{Fe}_2\text{O}_3$ ratios but, in the case of kyanite-free rocks, a narrow range of $\text{FeO}:\text{MgO}$ ratios. An exception is sample 2A which has a very high $\text{FeO}:\text{MgO}$ ratio because of a high FeO content.

Kyanite bearing schists have highly variable $\text{FeO}:\text{MgO}$ ratios. K_2O content exhibits a tendency to be slightly higher in kyanite bearing rocks than in those free of kyanite (Fig. 9). From Fig. 11 it can be seen that an increase in the $\text{Al}_2\text{O}_3:\text{MgO}$ ratio is accompanied by an increase in the $\text{Fe}_2\text{O}_3:\text{FeO}$ ratio. Those samples richest in Al_2O_3 and Fe_2O_3 contain abundant kyanite.

Figure 9. Vector diagram illustrating compositional variations of analysed schists (mole percent). Kyanite bearing samples are represented by dashed vectors.

Figure 10. Ray diagrams showing the range of the ratios $\text{FeO}:\text{Fe}_2\text{O}_3$ and $\text{FeO}:\text{MgO}$.

Figure 11. Composition of the schists in terms of the five major oxides, the iron oxides being shown as auxiliary vectors.

MINERAL CHEMISTRY

It was not possible to satisfactorily separate the minerals from some of the rocks which were analysed chemically, because of their fine grain size or the abundance of inclusions. The separation and analytical techniques used are given in Appendices C and D.

Muscovite

The muscovites were not analysed chemically. Precession photographs of the muscovite from sample 9B show it to be the $2M_1$ polymorph. Basal spacings obtained using a diffractometer show that muscovite is the only white mica present.

Biotite

The chemical composition and structural formulae of the biotites (based on 20 oxygen atoms) are listed in Table 3. These have an unusually high FeO content and low Fe₂O₃ and TiO₂ contents as compared with biotites from other schists and gneisses (Deer, Howie and Zussman, 1962, 3, p 81). They also vary widely in FeO:MgO ratios considering their small areal distribution. The relation of the FeO:MgO and FeO:Fe₂O₃ ratios to the rock oxidation ratio (Chinner, 1960) is shown in Fig. 12 a and b. It is evident that as the oxidation ratio increases, the Mg and Fe⁺⁺⁺ in the biotite increase.

Garnet

The compositions of the garnets analysed are listed in Table 4. The structural formula of garnet 7B, the only concentrate sufficiently pure to warrant complete analysis, is:

The cell edges of the garnets were obtained from powder patterns taken on a Siemens 114.6 mm diameter camera with Straumanis film arrangement and corrected for shrinkage. Filtered iron radiation was used. The α_1 and α_2 lines of the 842; 664; 941; 10,4,0; 10,4,2; and 880 reflections were measured.

The garnets are compared with some published analyses of other garnets coexisting with staurolite in a four component vector diagram (Fig. 13). It is apparent that the almandine and spessartine contents vary widely whereas the pyrope and grossular contents are relatively constant. Of the garnets plotted 30 of the 33 lie in the range 11^{+4} mole percent pyrope and $10^{+4}\frac{1}{2}$ mole percent grossular. The almandine content ranges from 50-85 mole percent, the amount being related to the oxidation ratio of the rock (Fig. 12b and c). This relation is at variance with that proposed by Sturt (1962), which is that the CaO and MnO content of a garnet is related to the metamorphic grade.

Staurolite

The compositions of the analysed staurolites are entered in Table 5. Because of the refractory nature of the mineral, determination of FeO and H₂O was not attempted.

In order to obtain some measure of the range of substitution of Mg and Mn for Fe in staurolite the compositions of 35 staurolites, obtained from the literature (Juurinen 1956; Deer, Howie and Zussman, 1962, 1 ; Card 1964), were plotted on a three component triangle with total iron, magnesium and manganese as compositional variables (Fig. 14). The compositional range is shown by the shaded area in the triangle and the points represent the compositions of the staurolite analysed during this investigation. The manganese content of all samples is less than 5 mole percent whereas the magnesium content varies from 15-35 mole percent.

As the unit cell of staurolite contains 2 formula units (Deer, Howie and Zussman 1962, 1) there are four (Fe, Mg) sites. It is possible that three of these are preferentially occupied by Fe.

Fig. 12b shows that the Mg:Fe ratio in staurolite, as in the case of the other ferromagnesian minerals, correlates positively with rock oxidation ratio.

Table 3
Biotite analyses

	1	2A	7A	7B	7C	8	9B	10
SiO ₂	-	35.2	38.7	36.7	37.8	36.1	36.1	36.7
Al ₂ O ₃	-	19.2	19.4	20.7	19.2	19.3	19.9	19.6
Fe ₂ O ₃	2.1	2.1	2.4	2.5	2.6	2.3	1.8	1.5
FeO	12.3	20.6	10.3	9.9	11.3	14.7	15.4	14.9
MgO	12.6	7.6	14.1	15.2	14.0	12.1	11.5	11.3
CaO	0.16	0.08	0.08	0.07	0.08	0.11	0.08	0.08
Na ₂ O	0.28	0.24	0.48	0.36	0.40	0.50	0.28	0.32
K ₂ O	8.3	8.8	7.9	8.4	8.5	8.5	8.7	9.0
TiO ₂	1.8	1.6	1.4	1.3	1.4	1.4	1.6	1.8
MnO	0.15	0.01	0.08	0.19	0.10	0.09	0.04	0.09
P ₂ O ₅	0.01	0.03	0.04	0.01	0.03	0.05	0.02	0.02

Molecular formulae based on 20 oxygen atoms

Si	4.91	5.10	4.84	5.02	4.90	4.89	4.98
Al	3.15	3.01	3.22	3.00	3.08	3.17	3.11
Σ z	8.06	8.11	8.06	8.02	7.98	8.06	8.09
Fe ⁺⁺⁺	0.22	0.24	0.25	0.26	0.23	0.18	0.15
Fe ⁺⁺	2.40	1.13	1.09	1.25	1.67	1.74	1.68
Mg	1.57	2.77	2.99	2.77	2.45	2.32	2.27
Mn	0.00	0.01	0.02	0.01	0.01	0.01	0.01
Ti	0.17	0.14	0.13	0.14	0.15	0.16	0.19
Σ y	4.36	4.29	4.48	4.43	4.51	4.41	4.30
Na	0.07	0.12	0.10	0.10	0.13	0.07	0.08
K	1.56	1.33	1.41	1.44	1.47	1.50	1.56
Ca	0.01	0.01	0.01	0.01	0.02	0.01	0.01
Σ x	1.64	1.46	1.52	1.55	1.62	1.58	1.65

Table 4
Garnet analyses

	2A	7A	7B	7C	8	9B	10
SiO ₂	-	-	37.5	-	-	-	-
Al ₂ O ₃	19.8	19.6	22.8	-	-	18.6	15.8
Fe ₂ O ₃	0.0	0.0	0.0	-	0.0	0.0	0.0
FeO	30.5	19.7	21.0	17.1	18.1	24.2	17.6
MgO	1.3	2.6	3.5	2.4	1.7	2.3	1.9
CaO	1.8	2.8	3.2	5.5	5.9	2.6	1.8
MnO	0.13	3.0	10.9	5.5	2.9	2.0	3.3
TiO ₂	0.20	0.60	0.14	0.09	0.58	0.08	0.06
Alm.	86.6	63.7	49.5	50.2	57.3	72.0	66.0
Py.	6.5	14.9	14.7	12.7	9.5	12.2	12.7
Gross.	6.5	11.6	9.7	20.7	23.7	9.8	8.6
Spess.	0.4	9.8	26.1	16.4	9.3	6.0	12.7
a, Å	11.54	11.57	11.57	11.57	11.55	11.55	11.56

Table 5
Staurolite analyses

	2A	7A	7B	7C	8	9B	10
SiO ₂	30.3	-	27.8	29.3	27.3	28.1	-
Al ₂ O ₃	53.1	-	55.4	54.7	54.9	55.1	-
FeO	14.4	10.3	11.8	12.2	13.9	13.6	9.9
MgO	1.2	2.5	3.1	2.7	2.3	2.0	1.2
CaO	0.04	0.07	0.06	0.02	0.08	0.01	-
MnO	0.01	0.38	0.81	0.46	0.30	0.13	-
TiO ₂	0.51	1.9	0.60	0.52	0.61	0.69	0.59
Total	99.56	-	99.57	99.90	99.39	99.63	-

Figure 12. Correlation of rock oxidation ratio with
 (a) biotite oxidation ratio, (b) mineral total iron:
 magnesium ratio, (c) molar ratio of iron in garnet.

Figure 13. Four component vector diagram showing the composition of garnet coexisting with staurolite.

Figure 14. Range in composition of 41 staurolites; those analysed during this study are represented as dots.

DISCUSSION OF RESULTS

Evidence for equilibrium

In the rocks studied textural evidence of chemical disequilibrium, such as reaction rims or pseudomorphs, is absent. Accordingly, as tests of equilibrium, resort must be made to:

- (a) the phase rule, and
- (b) the distribution of elements between coexisting minerals.

The phase rule

If a mineral assemblage is considered to be a thermodynamic system at equilibrium, then:

$$F = C - P + 2 \text{ (Turner \& Verhoogen, 1960, p. 497)}$$

where

F is the variance or number of degrees of freedom

P is the number of phases

C is the minimum number of independent components, in terms of which may be defined all the mineral phases observed in the rock being studied.

At independently fixed temperature and pressure,

$$P \leq C \quad (\text{Goldschmidt's "Mineralogical phase rule"})$$

In order to derive the variance of the system as defined above, the phases were listed and the major components necessary to define each phase were deduced.

Water was regarded as being "in excess" during metamorphism, that is, present as a phase approximating pure water.

The hematite-ilmenite intergrowths were considered to have exsolved from one phase, and in the garnets MnO and CaO were considered to be components. In the case of garnet from sample 2A, which contains only trace amounts of MnO, only CaO was considered.

The calculation of components in sample 8 is shown in Table 6.

The staurolite schists (except sample 2A) contain 9 phases and 11 components, that is $F = 4$.

Sample 2A contains 7 phases and 8 components, that is $F = 3$.

The kyanite-staurolite schists contain 10 phases and 11 components, that is $F = 3$.

Sample 6, the chlorite-kyanite schist contains 7 phases and 8 components, that is $F = 3$.

In all the above assemblages therefore, $P \leq C$, indicating no evidence of disequilibrium.

Distribution relations

One of the conditions for equilibrium is that the chemical potential of a component be the same in all phases. The relationship between the atomic ratio of

an element in a mineral and that in a coexisting mineral may be shown diagrammatically. This type of diagram was introduced by Roozeboom (1891) and later used extensively by Kretz (1959, 1960, 1961, 1964). The curves obtained are referred to as distribution curves, each point representing a mineral pair. Provided all points plotted on such a diagram represent minerals having formed under essentially the same conditions then a smooth curve should result if equilibrium was established.

The theoretical basis and the interpretation of these diagrams, particularly applied to geology, are given in papers by Kretz, Bartholome' (1962) and summarized by McIntire (1963). Because of the differences between distribution formulae arrived at by Kretz and by Bartholome' an attempt is made here to correlate them.

Kretz (1961) showed that under equilibrium conditions for coexisting phases $\alpha = (A,B)M$ and $\beta = (A,B)N$, where A,B,M and N are chemical species, the distribution coefficient, K_D , with reference to species A:

$$K_D = \frac{x_A^\alpha (1-x_A^\beta)}{(1-x_A^\alpha) x_A^\beta} = K_f \cdot \exp.(\Delta G/(RT)) \dots\dots\dots (1)$$

where

x_A is the mole fraction of A, equal to $A/(A+B)$

K_f is the value depending on the activity coefficients of chemical species A and B in phases α and β .

ΔG is the Gibbs free energy change of the reaction:

Table 6
Calculation of components
Sample 8

Phases	Components										
	SiO2	Al2O3	K2O	MgO	FeO	MnO	CaO	Na2O	Fe2O3	TiO2	H2O
quartz	+										+
muscovite	+	+	+								+
biotite	+	+	+	+	+	+					
garnet	+	+		+	+		+				
staurolite	+	+		+	+						+
plagioclase	+	+					+	+			
magnetite					+				+	+	
hemo-ilmenite											+
vapour											

Total: 9 phases; 10 components

T is the absolute temperature

R is the gas constant

If the mixture is ideal then $K_f = 1$, that is, at fixed temperature and pressure, K_D is a constant.

Bartholome' (1962) has shown that the iron-magnesium ratios in coexisting olivines and calcium-poor pyroxenes may be related by the expression:

$$\left(\frac{\text{Fe}^{++}}{\text{Mg}} \right)_{\text{ol}} = K \left(\frac{\text{Fe}^{++}}{\text{Mg}} \right)_{\text{Ca-P}}^x$$

where K is a temperature dependent function.

The general form of this equation may be written:

$$\frac{A^Q}{B^Q} = K \left(\frac{A^B}{B^B} \right)^x \dots\dots\dots (2)$$

where for ideal solutions, $x = 1$

Further if $X_A = A/(A + B)$

then, $A/B = X_A/(1-X_A)$

Thus equation (2) becomes

$$\left(\frac{X_A^Q}{1-X_A^Q} \right) = K \left(\frac{X_A^B}{1-X_A^B} \right)^x$$

$$\text{or } \left(\frac{X_A^Q}{1-X_A^Q} \right) \cdot \left(\frac{1-X_A^B}{X_A^B} \right) = K \left(\frac{X_A^B}{1-X_A^B} \right)^{x-1}$$

Combining this with (1):

$$K_D = K \left(\frac{X_A^B}{1-X_A^B} \right)^{x-1} = K_f \cdot \exp.(\Delta G/(RT)) \dots\dots (3)$$

For ideal solutions, $K_f = 1$ and $x = 1$

therefore $K = \exp. (\Delta G/(RT))$

A possibility therefore exists that in the general case

$$K_f = \left(\frac{x_A^B}{1-x_A^B} \right)^{x-1} \dots\dots\dots (4)$$

An equation similar to (4) was obtained empirically by Hill, Durham and Ricci (1940) who investigated the equilibrium relations between solid solutions of isomorphous salts and their saturated aqueous solutions.

The distribution relations of Fe^{++} , Mg and Mn, octahedrally coordinated elements, among coexisting garnet, biotite and staurolite were determined. The term "molar ratio" used in the following discussion, is defined as the molar concentration of an element in a particular site in a crystal structure, that is the molar ratio of Mg equals:

$$X_{Mg} \text{ (garnet)} = Mg / (Mg + Fe + Mn + Ca)$$

$$X_{Mg} \text{ (biotite and staurolite)} = Mg / (Mg + Fe + Mn)$$

The distribution curves obtained are shown in Figures 15 & 16. Total iron content of each mineral has been used in all calculations. The distribution coefficient was calculated for each mineral pair using equation (1), the results averaged and the curve plotted using this equation.

The distribution of Mg and Mn is almost constant for each mineral pair. The two points on the diagrams of Mn distribution which appear erratic are probably caused by incorrect Mn determinations on biotite 7C and

Figure 15. Distribution plots of Fe and Mg between garnet, biotite and staurolite.

$$X_{Mn} = Mn / Fe + Mg + Mn + Ca$$

Figure 16. Distribution plots of Mn between garnet, biotite and staurolite.

staurolite 7A. The large scatter on two of the iron distribution diagrams is probably the effect of Ca and Mn in the garnets on the distribution coefficient (Kretz, 1959). The correlation obtained for Mg is a strong argument for equilibrium having been attained, at least with respect to the distribution of this element. Accordingly the temperature of equilibrium was either constant for the suite or, what is more likely, the distribution coefficient is relatively insensitive to the range of temperature represented.

The total iron : magnesium ratios of biotite and staurolite were plotted on log-log paper (Fig. 17). The distribution closely approximates a straight line, the equation for the line of best fit being:

$$K_D = 5.38 \left(\frac{x_{Mg}^S}{1-x_{Mg}^S} \right)^{0.15}$$

This indicates that the distribution of iron and magnesium between biotite and staurolite departs slightly from ideality. In equation (3) p. 47, $x = 1.15$, compared to 1.00 for ideal solutions. If Mn is included in the molar ratios the slope and intercept of the line are changed but slightly and the scatter is slightly greater.

Representation of phase assemblages

Silicate minerals

Garnet, staurolite, and biotite were plotted on the AFM face of a AKFM tetrahedron by projecting through

Figure 17. Distribution of Fe and Mg between biotite and staurolite plotted on log-log paper.

Figure 18. Compositions of minerals coexisting with muscovite and quartz, shown on the AFM face of a AKFM tetrahedron by projecting through muscovite.

muscovite (Thompson, 1957). The coexisting minerals are joined by tie lines (Fig. 18). Where alumina was not determined the tie lines are shown dotted.

The alumina in the garnet was corrected for MnO and CaO present by subtracting the alumina necessary to form spessartine and grossular respectively, as neither of these end members are shown on the projection. The alumina in the staurolite was corrected in a similar manner. Na₂O was added to the K₂O in the biotites; total iron was plotted as FeO in all cases.

It was not possible to plot the rock composition on this diagram because of the large content of opaque minerals and the difficulty of obtaining an accurate estimate of FeO in them.

Considering first the staurolite-garnet-biotite schists it was noted earlier (p.44) that they have four degrees of freedom, except for sample 2A which has three. It is reasonable to assume that temperature, pressure and the molar ratio of ferrous iron correspond to three degrees of freedom. The remaining degree of freedom could be the mole fraction of manganese. This assumption is consistent with the absence of manganese from sample 2A in which there are three degrees of freedom.

The kyanite-staurolite-garnet-biotite schists have only one compositional degree of freedom if temperature and pressure are independently variable. However, the

latter assumption may not hold for sample 10 which was sampled from the eastern limit of the staurolite zone. In thin section the staurolite in this sample consists of small irregular grains. This evidence suggests that sample 10 lies on an isograd and thus the temperature and pressure may not have been independently variable.

The remaining two analysed rocks containing kyanite with staurolite, (7A and B), contain garnets with very similar Ca and Mg content but widely different Mn and Fe content. Because Fe and Mn vary reciprocally, they may not be independently variable. There is a definite possibility that the mole fraction of Mn may be the independent compositional variable; that is, the presence of sufficient Mn stabilized the garnet in this assemblage.

The silicate mineral assemblages studied may be represented schematically on an AFM diagram (Fig. 19), provided it is realized that the molar fraction of Mn is an important compositional variable, not included in the diagram, and that garnet may be present as a result in assemblages from which it is excluded in the projection.

Opaque minerals

Composition-paragenesis diagrams for the systems Fe-O-S (Kullerød, 1957) and Fe-O-TiO₂ (Buddington, 1963, 1964) were used to construct an activity diagram for the system Fe-O-S-TiO₂ (Korzhinskii, 1959, p.90, 91). This diagram is drawn in isometric projection (Fig. 20). The

Assemblages include

Quartz

Muscovite

Figure 19. Diagrammatic representation of mineral assemblages observed during this study, those encountered being shown by dots.

Figure 20. Schematic isometric projection of the activity diagram of the system Fe-S-O-TiO₂. Two mineral assemblages occur along surfaces; those including ilmenite board the ilmenite field, which is left open for clarity.

Figure 21. Activity diagram of the system Fe-O-S-TiO₂ showing all the phases in equilibrium with ilmenite. Observed assemblages are shown schematically by dots.

temperature chosen is below that of the stability field of wüstite (570°C at one atmosphere total pressure). The diagram was simplified by assuming no ilmenite-hematite solid solution.

As all the schists examined contain ilmenite, a projection of the surface in the activity diagram of the ilmenite field, on to the a_S - a_{O_2} plane was drawn (Fig. 21). All phases in equilibrium with ilmenite can thus be shown on a two dimensional diagram. It should be noted that all assemblages in or bordering the ilmenite-hematite field contain ilmenite-hematite intergrowths.

The assemblages observed are shown schematically on the diagram by points grouped as closely as the geometry permits. The most reduced assemblage encountered: pyrite-pyrrhotite-ilmenite (without hematite intergrowths), occurs in the graphitic schists; the stability limit of graphite is illustrated schematically in Fig. 21. In order to explain the other assemblages the only major variable that need be considered is the activity or partial pressure of oxygen, because the activities of sulphur and titania could have been virtually constant among the samples.

Control of mineral paragenesis

Metamorphic grade

There is no consistent relation between iron-magnesium ratios of the minerals and geographic distribution

of samples (Fig. 18). However, there are significant differences in mineral iron-magnesium ratios among samples only a few tens of feet apart. This leads to the conclusion that the effect of temperature and pressure is insignificant compared to other factors. This conclusion is further supported by the almost constant distribution coefficients presented earlier.

Rock composition

Rock oxidation ratio (Chinner, 1960) is a convenient measure at constant temperature and total pressure, of partial oxygen pressure during metamorphism, and it has been shown that this ratio correlates with iron-magnesium ratios in the minerals (Fig. 12). The effect of increasing oxidation ratio is to reduce the amount of iron available to silicate minerals, because they may accommodate little ferric iron. The higher the oxidation ratio, the greater the ratio of magnesium to iron in the ferromagnesian minerals. Oxidation of iron effectively shifts the bulk composition of the rock away from the FeO corner of the triangle of Fig. 18, thus leading to assemblages containing relatively aluminous or magnesian minerals. The samples with highest oxidation ratios (samples 6 & 2B) contain the pair chlorite-kyanite. Accordingly the oxidation ratio and $\sum R_2O_3 : \sum RO$, which is in part consequent on bulk composition of the original sediment, and

partly a function of oxidation ratio, are the important variables affecting the mineral paragenesis.

Origin of the different oxidation ratios

As the oxidation ratios vary significantly over distances of a few tens of feet (samples 7A, B, C) they may be a function purely of the state of oxidation of the sediment prior to metamorphism. The amount of carbon in the original sediment must also be considered. If the carbon present originally in the rock was completely oxidized during or before metamorphism, and the system closed to oxygen and hydrogen, then any oxidation ratio might have resulted, dependent only on the original oxidation ratio and the original carbon content.

Miyashiro (1964) proposed that the aqueous fluid present during metamorphic processes must normally contain hydrogen in excess of that in pure water, produced in most cases by the dissociation of water and equilibration with graphite. That is, the following reactions are combined:

Miyashiro suggested that H_2 may move from graphite-bearing rocks to nearby graphite-free ones.

In the area studied, bands of graphitic schists alternate with graphite-free schists. The origin of the

different oxidation ratios may thus lie in limited migration of hydrogen from the graphitic schists as suggested by Miyashiro. Extensive migration, however, would homogenize the oxidation ratios.

CONCLUSIONS

1. Phase rule considerations and distribution relations indicate a high probability that the rocks studied attained local equilibrium during metamorphism, the partial pressure of oxygen varying among sample localities.
2. The FeO:MgO ratios of the ferromagnesian minerals are primarily dependent on the oxidation ratio of the rock, that is the availability of ferrous iron to the silicates. Within the limits of the grade of metamorphism studied they are relatively insensitive to temperature and pressure, or the temperature range is small. The ferrous-ferric ratio of biotite is also dependent on the rock oxidation ratio.
3. The opaque mineral assemblage is for the most part controlled by partial oxygen pressure, the activities of sulphur and titania playing a minor part.
4. Compositional variables affecting the silicate mineral assemblage are $\sum R_2O_3 : \sum RO$ ratio and the concentration of manganese. Aluminous, highly oxidized rocks contain kyanite and/or chlorite. Garnet may be stabilized with kyanite by relatively high MnO content. The size of the garnet crystals decreases with increasing MnO.
5. The distribution of Fe and Mg between biotite and staurolite is not ideal, but is related to the mole

fraction of Mg in staurolite, the relationship being:

$$K_D = 5.28 \left[x_{Mg}^S / (1 - x_{Mg}^S) \right]^{0.15}$$

6. Staurolite usually contains less than 5 mole % Mn, whereas the Mg content varies from 15-35 mole %.

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APPENDICES

Appendix A

Field numbers and localities of samples.

Sample No.	Field No.	Locality
1	G-5-61	Hwy 506, 1.5 mile S.W. of Marble Lake.
2A	G-9-61	Junction of Hwy 506 and power line, S.W. corner of Marble Lake.
2B	G-69-61	
3A	G-44-61	Power line cut, S. shore of Georgia Lake, 500 ft. W. of outlet.
3B	G-103-62A	Outlet of Marble Lake, under bridge on Hwy 506.
3C	G-103-62C	
4	G-68A-61	E. side of Green Bay, Mississagagon Lake, 700 ft N. of Hwy 506.
5	G-60-62	Power line cut, 100 ft N. of Hwy 506, 1500 ft W. of Twin Pines road, S. of Mississagagon Lake.
6	G-10-61A	Power line cut, 100 ft N. of Hwy 506 at Flieler's Flying Service road, S. of Mississagagon Lake.
7A	G-78A-61	E. tip of Mississagagon Lake, 300 ft N. of outlet creek at dam.
7B	G-20-61	
7C	MD-41A-60	
7D	MD-41C-60	
8	MD-25X-60	Power line at access road, 0.3 mile N. of Fernleigh.
9A	MD-43A-60	Kleedorfer farm road, 0.4 mile N. of Hwy 506, E. end of Schwager's Lake.
9B	MD-43D-60	
10	G-26-61	Ridge, 0.2 mile N. of junction of Hwy 506 and 509, 1 mile W. of Ardoch.

Appendix B

Derivation of a five component vector diagram from a five component bar graph

Appendix C

Sample preparation

Comminution of samples.

The unweathered rock sample weighing approximately 2-6 lbs. was broken into $1\frac{1}{2}$ inch pieces with a hydraulic splitter or hammer and plate, and crushed with a Chipmunk jaw crusher to obtain a product no greater than $\frac{1}{4}$ inch. The product was split with a sample splitter (or by coning and quartering) to give a representative 10 gm fraction for bulk analysis. This fraction was loaded into ceramic capsules with one ceramic ball, shaken for 10 minutes in a Pica Blender Mill and screened. The process was repeated until all the sample was less than 100 mesh. Crushing was finished with mullite mortar when necessary.

The remainder of the jaw crusher product was repeatedly passed through a Bico pulverizer with ceramic plates, starting with a $1/16$ inch gap which was gradually reduced until most of the sample passed through a 100 mesh screen. The + 100 mesh fraction was then crushed to less than 100 mesh using a mullite bucking board. The combined - 100 mesh fraction was screened through 150, 200, 270, and 325 mesh screens using an automatic shaker. Each fraction was then washed with water to remove dust.

Separation of minerals.

Because of the abundance of inclusions in the minerals studied most of the separations were performed using

the -200 + 270 and the -270 + 325 mesh fractions. A generalized flow sheet for the -270 + 325 mesh fraction is shown in Figure 22.

The principles of optimum operation for the several mineral separation techniques are summarized below.

Isodynamic separator.

A Cooke isodynamic separator (similar to the Franz) was used. The maximum current was selected at which the grains did not stick to the magnet; the side slope was then adjusted to obtain the required separation, the current being reduced when the side slope became so large that the grains spilled over the side of the chute. The forward slope chosen was dependent on the grain size and the speed of feed. For more detailed information see McAndrew (1957).

Elutriating tube.

An elutriator modelled after that of Frost (1959) was used in a few cases for concentrating the heavier minerals in the presence of a large amount of biotite and for floating off biotite when present in small amounts relative to the other minerals.

Heavy liquids.

For separating a relatively large amount of material in bromoform or methylene iodide 125 ml Squibb separating funnels with Teflon plugs were used in an International centrifuge having the appropriate carrier.

Figure 22. Flow chart for separating constituent minerals from the -270+325 mesh fraction.

When the amount of sample was small a 15 ml polypropylene centrifuge tube with a conical bottom was used. This tube was extremely useful when using Clerici solution as the heavier fraction stuck to the bottom of the tube and the liquid and lighter fraction could be poured off without any auxiliary apparatus. This procedure is not satisfactory if the grain size is greater than 200 mesh.

During and after the physical separation procedures described above the mineral separates were carefully observed under both binocular and polarizing microscopes. Particular attention was paid to the possibility of intermixture of garnet and staurolite.

The kyanite and opaque minerals in the analysed samples were positively identified by powder x-ray diffraction photographs.

Final treatment of minerals before analysis.

The biotite samples and the garnet and staurolite from sample 7B (Plate 7) were considered satisfactorily pure and free from inclusions after the above separation procedure. Most of the garnet and staurolite concentrates, even in the -270 mesh fraction contained abundant inclusions of quartz and opaque minerals. Before analysis the garnets were ground under acetone by hand in a mullite mortar for 15 minutes, and the staurolites under

alcohol in an automatic mullite mortar for 1 - 2 hours. After drying under a heat lamp the samples containing opaque inclusions were transferred to 150 ml beakers to which 50 ml of 1:1 HCl was added. These were covered and placed on top of an oven (at approximately 50° C) for several days. The powders were then filtered off, washed well with water and dried. This treatment, while not generally recommended, was considered to be the only satisfactory means of purification in this case. Because both minerals are refractory, it is considered that the chemical composition was unaffected by this treatment.

Plate 7. Samples of analysed staurolite (A), biotite (B) and garnet (C), from sample 7B.

Appendix D

Chemical analysis

Basic procedure.

The analytical scheme is shown diagrammatically in Figure 23.

Solution A is used for the determination of silica and alumina. It is prepared by fusing 50 mg of the sample or standard (NBS Soda feldspar No. 99) with NaOH in a nickel crucible at dull red heat, followed by solution of the melt in dilute HCl and dilution to one litre. (Shapiro and Brannock, 1956).

Silica is determined by reducing the yellow silicomolybdate complex (in a solution which is 2N with respect to H_2SO_4 in order to prevent the interference of phosphorus) to molybdenum blue with stannous chloride and measuring the absorbance of this solution at 825 mu. This method is adapted from that of Milton (1951) by J. M. Moore.

Alumina is determined by measuring the absorbance at 475 mu of a solution containing the complex of Al, Ca and Alizarin red-S buffered at pH 4.5. Iron is complexed with thioglycollic acid. (Based on the procedure of Shapiro and Brannock, 1956).

Figure 23. Basic scheme of chemical analysis.

Solution B is used for the determination of CaO, MgO, total iron, Na₂O, K₂O, MnO, TiO₂ and P₂O₅. It is prepared by dissolving 400 mg of sample in a mixture of HF, H₂SO₄ and HNO₃ in a 150 ml Teflon beaker. The HF and HNO₃ are removed by heating on a hot plate until the H₂SO₄ is fuming. The residue is cooled, dissolved in water, diluted to 200 ml and transferred to a polyethylene bottle. (Based on the method of Shapiro and Brannock, 1956).

Calcium is determined by titration with EDTA at pH 13, using a modified Calcein indicator. Al, Fe and Mn, which would otherwise interfere are complexed with triethanolamine. The pH is adjusted with KOH rather than NaOH as the presence of large quantities of sodium gives rise to a green fluorescence which obscures the end point. The use of a magnetic stirrer is mandatory for all EDTA titrations if a sharp reproducible end point is to be obtained. (Based on a procedure of C. E. S. Davis, Division of Cements and Refractories, C.S.I.R.O. Melbourne, personal communication).

Calcium plus magnesium is determined by titration with EDTA at pH 10 using screened o-cresolphthalein complexone indicator. Al, Fe and small amounts of Mn which would otherwise interfere are complexed with triethanolamine. (C. E. S. Davis, personal communication).

Magnesium is determined by difference.

Total iron is determined by measuring the absorbance of the orange ferrous-orthophenanthroline complex at 506 mu in a solution of pH 4.5 (acetate buffer). (Based on the procedure of Shapiro and Brannock, 1956).

Manganese is determined by oxidizing Mn to permanganate with potassium periodate and measuring the absorbance of the solution at 525 mu. (Shapiro and Brannock, 1956).

Titanium is determined by measuring the absorbance of its Tiron complex at 380 mu. The solution is buffered at pH 3.8 and thioglycollic acid is used to complex the iron which would otherwise interfere. (Rigg and Wagenbauer, 1961).

Phosphorus is determined by reducing the yellow phosphomolybdate complex to "molybdenum blue" with ascorbic acid and measuring the absorbance at 825 mu. (Riley, 1958).

Sodium and potassium are determined on a diluted aliquot of solution using an Eel flame photometer with Propane-air flame and sodium and potassium standards of appropriate concentration.

Ferrous iron is determined on a separate sample by decomposing at room temperature in a HF solution containing a known amount of quinquivalent vanadium which oxidizes the ferrous iron as it passes into solution. After standing until decomposition is complete the excess V^{+5} is reduced by a known quantity of ferrous ammonium sulphate, the excess of the latter being titrated with standard $K_2Cr_2O_7$ using barium diphenylamine sulphonate indicator. Boric acid is used to complex the HF, and H_3PO_4 to complex the ferric iron formed. (After Wilson, 1955, 1960).

Ferric iron is determined by difference.

Water and other volatiles are determined by loss on ignition at $900-1000^{\circ}C$. A correction for the oxidation of ferrous iron is made on the basis of an FeO determination after heating.

Modification for garnets high in MnO.

Solution A.

The permanganate ion forms during solution of the sample and must be reduced by adding a few drops of dilute ferrous chloride solution.

Solution B.

MnO precipitates during the fuming process but may be redissolved by the addition of a few crystals of sodium sulphite to the aqueous solution of the fumed residue and boiling while still in the Teflon beakers. The alkalies can not then be determined on this solution but this is unnecessary in the case of garnets.

Magnesium could not be determined by the usual procedure due to the interference of manganese. It was determined by measuring the absorbance at 545 mμ of the Clayton-yellow-magnesium lake formed in strong alkali solution. KCN and triethanolamine are used to complex interfering elements and polyvinyl alcohol and aluminium nitrate to stabilize the colour. (Meyrowitz, 1964).
Modification for staurolite and rocks containing staurolite.

B solution.

As staurolite does not dissolve in HF and H₂SO₄ the sample is fused with NaOH in a silver crucible at 600 - 700°C (Riley, 1958). The melt is then dissolved in 1:1 HCl, transferred to a Teflon beaker, evaporated to near dryness and digested overnight in HF and H₂SO₄. This is then evaporated to H₂SO₄ fumes, cooled, dissolved in water and diluted to volume. The solution is filtered through dry filter paper to remove the precipitated silver salts.

Table 7

Replication of chemical analyses.

	Rock	biotite	garnet	staurolite
SiO ₂	3-2	1-2	1-2	1-2
Al ₂ O ₃	3-2	1-2	1-2	2-2
FeO*	1-2	1-2	1-2	1-2
FeO	2-1	2-1	2-1	2-1
MgO	1-1	1-1	1-2	1-1
CaO	1-1	1-1	1-1	1-1
Na ₂ O	1-3	1-3	-	-
K ₂ O	1-3	1-3	-	-
TiO ₂	1-1	1-1	1-1	1-1
MnO	1-1	1-1	1-2	1-1
P ₂ O ₅	1-1	1-1	-	-

Legend: Number of sample solutions - number
of determinations on each sample
solution.

* Total iron as FeO.

Ferrous iron was not determined on the staurolites due to the difficulty of dissolving the mineral in an inert atmosphere.

Ferric iron in the staurolite bearing rocks was determined by difference from total iron and FeO values using HF-H₂SO₄ solutions only. Because staurolite is not dissolved the determination is in error by the amount of Fe₂O₃ present in the staurolite. This could at the most introduce a negative absolute error of 0.5% Fe₂O₃, which was considered sufficiently small to neglect in this investigation. FeO values for the rock samples are determined by difference between Fe₂O₃ determined in this way and total iron determined on the solution obtained from NaOH fusion.

Replication

The number of sample solutions prepared, and the number of determinations of each constituent in each solution are shown in Table 7.

Precision and accuracy

Analyses of the reference rock standards Syenite Rock-1, W-1 and Msusule Tonalite-1, which were performed under the same conditions, with the same care and at the same time as the other analyses presented in this paper, are listed in Table 8. These figures, together with the

correlations and structural formulae presented in the text indicate the reliability of the results obtained.

The only analytical procedure employed which has not yet been widely used is the Clayton yellow magnesium method. Duplicate analyses of the same sample solution by this technique, performed two months apart are listed in Table 9.

Table 8

Analyses of Reference Rock Standards (weight %)

Sample	W - 1		SR - 1	(b)	T1	
		(a)				(c)
SiO ₂	53.0*, 52.8*, 52.5*	52.54	60.4*, 60.0*	59.3	63.6	62.6
Al ₂ O ₃	15.3*, 15.2*, 15.2*	15.05	9.5*, 9.1*	9.3	16.6	16.5
FeO	9.7, 9.7, 9.8	10.02	7.1, 7.2	8.0	5.2	5.43
FeO	8.6, 8.7	8.75	6.4, 6.1	6.08	2.9	2.90
MgO	7.0, 6.7	6.64	4.5, 4.3	4.02	1.8	1.89
CaO	10.7, 11.0, 10.8	10.98	10.0, 10.1	10.24	5.0	5.19
Na ₂ O	2.2, 2.2, 2.2	2.07	3.4, 3.4	2.99	4.2	4.39
K ₂ O	0.64, 0.64, 0.64	0.64	2.6, 2.5	2.71	1.2	1.23
TiO ₂	1.1, 1.1, 1.1	1.07	0.49, 0.46	0.52	0.56	0.59
MnO	0.18, 0.18, 0.18	0.16	0.42, 0.41	0.42	0.12	0.11
P ₂ O ₅	0.13, 0.13, 0.13	0.14	0.21, 0.21	0.19	0.15	0.14

* Analysed by R. Levick using methods described
in this paper.

(a) USGS Bull. 1113, arithmetic mean of preferred
values

(b) Report of nonmetallic standards committee, CAAS,
Applied Spectroscopy 15, No.6, 1961, arithmetic
mean.

(c) Standard geochemical sample T-1, Msusule tona-
lite, Supplement No. 1, 1963, Tanganyika Geolo-
gical Survey, arithmetic mean of preferred values.

Table 9

Determination of MgO by the Clayton yellow procedure

Date Per-formed	Garnets							T-1	W-1	SR-1
August	1.31	3.44	1.70	1.82	2.81	2.21	2.61	1.58	6.62	4.07
October	1.23	3.53	1.67	1.89	2.53	2.31	2.51	1.52	-	-