Pyroxenes, amphibole, and mica from the Tiree marble.[1]

(With Plate XIV.)

By A. F. HALLIMOND, M.A., Sc.D.

With chemical analyses by C. O. HARVEY, B.Sc., A.R.C.S.

[Read March 27, 1947.]

I. INTRODUCTION.

THE Tiree pink marble, a fine-grained, severely crushed limestone with evidence of earlier coarse crystallization, is exposed in several small areas up to 100 feet across on the farm of Balephetrish near the north coast of the island of Tiree in the Hebrides. It contains a remarkable quantity of dark silicate minerals and has discordant contacts with the adjacent Lewisian gneiss. The precise nature of its relation to the gneiss, and mode of emplacement, have been much discussed.[2] The writer has been permitted to consult accounts of the literature, by Mr. V. A. Eyles, and of the petrography, by Sir Edward B. Bailey, and is also indebted to those authors for discussion of the problems. At the suggestion of Sir Edward Bailey the present work was undertaken as a contribution to the study of this problem from the mineralogical point of view. Most of the determinations were made in 1938, but it was not possible to complete publication at that time.

Scattered somewhat evenly throughout the carbonate groundmass of the marble are numerous dark green crystalline patches which usually do not exceed half an inch across and are often much smaller; locally, however, there are pieces of dark gneiss-like rock of much larger size. The majority consist of pyroxene, sometimes accompanied by amphibole, &c., and the latter were regarded by Coomáraswámy and others as modified gneiss inclusions. Besides these assemblages the rock contains a subordinate amount belonging to other rock types, part of which may have been emplaced by the 'cold crush'; but many of these 'xenoliths', as is well known, are intergrown with uncrushed coarse calcite and must have been emplaced in the limestone at an earlier period, preceding the marmorization. For the present purpose attention

[1] Communicated by permission of the Director, Geological Survey and Museum.
[2] A. K. Coomáraswámy, Quart. Journ. Geol. Soc. London, 1903, vol. 59, p. 91.

has been confined to two hand-specimens of the predominant type, which were collected by Mr. V. A. Eyles; one representing the typical groundmass marble, with small scattered grains and groups of pyroxene; the other, a gneiss-like assemblage of pyroxene, amphibole, mica, and scapolite with coarse crystalline calcite. The dark minerals have been separated and their descriptions will be given below, with chemical analyses by Mr. C. O. Harvey. The silicates, except scapolite, are little altered, even the mica being apparently unaffected.

II. DESCRIPTION OF THE ANALYSED MATERIAL.

A. Pyroxene marble.

Locality: NE. end of shore section, Balephetrish, Tiree.

The hand-specimen shows dark pistachio-green pyroxene in a ground-mass of pink porcellanous calcite. The grains, which form single crystals or aggregates, usually less than $\frac{1}{8}$ of an inch across, are rounded and polished (coccolite), but often have incomplete faces of the forms a, b, m, and are elongated along the c-axis. Locally the aggregates are larger.

Microstructure.—The thin section S32453 shows much pyroxene with m cleavage and in places numerous twin-lamellae parallel to a, also parting cracks parallel to c. When calcite has been included, in embay-ments or isolated patches, it has rounded outlines. The pyroxene forms isolated grains or groups intergrown with calcite in a typical coarse polyhedral granular texture (pflaster) in which the pyroxene is sub-idiomorphic (pl. XIV, fig. 1). The chip shows that the section has passed close to the side of a large pyroxene aggregate, which has no doubt protected the calcite from being crushed. A duplicate section (pl. XIV, fig. 2) was cut through typical groundmass marble, and this shows rounded pyroxenes in crushed calcite; some aggregates contain pyroxene, apatite, and an ophitic mineral of moderate birefringence, uniaxial negative, refractive index below balsam. This resembles miz-zonite, but could not be completely identified and was not found in the tailing. Practically all the particles have their original outlines against coarse calcite in embayments, indicating that they were present before the 'cold crush'.

The first-mentioned section is crossed by a minute 'shatter-belt' of cleaved pyroxene fragments in a matrix of fine granular calcite. In places between the opened cleavages, and replacing the neighbouring calcite, is a minutely granular mineral of fairly low birefringence, refractive index 1·54, and lighter than bromoform. This is probably

chalcedony, introduced either at the time of the 'cold crush' or even later. There is no hornblende.

Separation.—The rock was crushed to pass 30 mesh to the inch and washed to remove fine dust. When stirred with dilute (1 in 5) acetic acid it effervesced freely. The insoluble part was dried and sieved, with further crushing and acid treatment. Finally the fraction 60–90 mesh was passed through a magnetic separator.[1] With full current the pyroxene was largely removed by the last magnet in a nearly pure state; the tailing consisted of small grains of pyroxene and a noticeable quantity of the granular silica, with a little apatite. After repeated magnetic treatment the concentrate was given a final wash with warm 1:4 hydrochloric acid and water and dried for analysis.

The density was determined in a specific gravity bottle, and the optical properties by means of the universal stage on a micro-section of the coarse separated grains mounted in plaster. β for sodium-light was measured on flakes showing an optic axis (nearly normal to the *c* parting) in a liquid mixture which was checked by the Abbe refractometer. The results are summarized in table I.

B. Gneiss-like aggregates in calcite.

Locality: Pink marble quarry, Balephetrish, Tiree.

The second hand-specimen represents a much less pure limestone, or calc-gneiss. Dark silicate grains up to $\frac{1}{3}$ of an inch across are scattered through fine-grained pale pink calcite, and towards one side there is an aggregate about 3 inches long of dark minerals mingled with calcite. Amphibole is present throughout along with more or less pyroxene, as polished grains which are often idiomorphic.

Microstructure.—The thin section S32451 and duplicates contain pyroxene, amphibole, biotite, altered scapolite, and calcite, with some apatite. The first two in places form composite aggregates up to 1 inch across. Calcite as included grains or embayments is coarsely crystalline and twinned, with rounded margins against the silicates, but the free calcite areas have been reduced to a fine granular condition. There is no chalcedony. The section shows a roughly banded arrangement, with pyroxene, or else amphibole and mica, predominating in different areas. The general texture of the aggregates is coarse polyhedral granular (pflaster) with subidiomorphic silicates (pl. XIV, figs. 4 and 5). The polyhedral boundaries are curved with a tendency to hexagonal outlines, a typical metamorphic texture which probably corresponds with pro-

[1] A. F. Hallimond, Min. Mag., 1930, vol. 22, pp. 377–381.

longed heating under conditions of physical and chemical equilibrium. In other places, however, the amphibole in particular has developed crystal outlines against the calcite and scapolite, or extends in irregular parallel growths often intergrown with biotite (pl. XIV, fig. 6). In places the outlines of the pyroxene suggest that it was being slowly absorbed while the amphibole and mica were increasing. The amphibole is occasionally in parallel growth with pyroxene, and the biotite often occurs with the basal plane parallel to the *a* face of the amphibole.

A distinctive arrangement of the minerals is the formation of narrow envelopes ('Schalen') surrounding the grains, the commonest example being a thin coating of scapolite surrounding amphibole in calcite (pl. XIV, fig. 3). The most probable cause seems to be a slow surface reaction under metamorphic conditions that led to the deposition of a single extended crystal; or the metamorphism of an existing altered surface. In pl. XIV, fig. 3 granules of pyroxene are included in the scapolite as if in a symplectite, but an envelope of pyroxene only was also seen, so this may indicate scapolitization of an original pyroxene envelope. Similar envelopes are an important feature in the Pargas limestone.[1]

The scapolite grains are nearly always polyhedral against silicates and calcite, and are probably not merely felspars replaced in situ. They are turbid from alteration to a micaceous mineral in parallel orientation. The bulk extinction and parting cracks are parallel with the elongation of the micaceous mineral, but the flakes are not all in one plane. Cleavage fragments in the tailing are roughly prismatic, and show a negative uniaxial interference-figure from areas of unaltered scapolite, emerging obliquely in the plane at right angles to the prism edges. This would agree with the development of the micaceous flakes along pyramidal planes of the scapolite, on which they confer a false cleavage. Both mica and scapolite have refractive index approx. 1·585 for vibrations parallel with the elongation of the flakes, i.e. approx. γ in each case. This corresponds with about 60 % Me in the scapolite, a normal limestone value. Mr. C. O. Harvey reports on the fragments: 'The mineral contains aluminium as a major constituent, a little calcium, and some magnesium, almost certainly less than 5 % MgO. Alkalis not determined.' Talc or phlogopite seems to be indicated.

Intergrowth of mica and pargasite.—P. Quensel[2] has described an example from Almunge in which lepidomelane forms 'long thin lamellae

[1] A. Laitakari, Bull. Comm. Géol. Finlande, 1921, vol. 10, no. 54. [M.A. 1–211.]
[2] P. Quensel, Bull. Geol. Inst. Upsala, 1914, vol. 12, p. 146.

parallel to the cleavage planes' in ferrohastingsite which has reaction rims of drop-like diopside. At Mansjö, also, the pargasite is often inter-grown with a dark phlogopite (table III); this mica is represented by the point 'new analysis 42' on the R_2O_3/RO diagram for the biotite group (p. 239, below) and is intermediate in composition between true phlogopite and the Tiree mica. Quensel seems to imply that the Almunge mica was parallel to the m faces of the amphibole, but measure-ments on the universal stage indicate that at Tiree it is orientated parallel to the face a, the trace of the cleavage being parallel in the two minerals (pl. XIV, fig. 6).

Separation.—Part of the dark aggregate, with the omission of some surface-weathered material, was crushed to −30 mesh and cleaned with acid in the same way as the preceding. Mica was removed by rolling the powder down an inclined surface and by electrostatic separation. The powder was reduced to 90–120 and 120–180 mesh, again cleaned with acid, and passed through the magnetic separator. The pyroxene was ultimately obtained fairly pure, but the amphibole contained biotite and particles of pyroxene with inclusions. The tailing consisted of altered scapolite, pyroxene, and some apatite, but no silica. The mica was rolled with a glass rod to break up some grains of altered scapolite that had lifted, and then sieved and washed. The pyroxene and amphi-bole were finally purified by floatation in mixtures of methylene iodide with benzol or aniline (see note at the end of this paper). Densities were determined as before and the data are summarized in table I.

III. CLASSIFICATION OF THE MINERALS.

The physical data agree with previous records for similar minerals, and the calculated atomic composition fulfils the requirements of the respective cell-formulae. When the present determinations were made, in 1938, it was difficult to complete the classification of the minerals because of the lack of summaries of the existing data. The new edition of Dana's 'System' is still in preparation, but in the interval several reviews of the mineral groups have been published, by means of which it has become easier to correlate the material. Reference will be made to these in the respective sections.

Diopside of normal marble (Lab. no. 1037).

The physical data and analyses indicate a diopside nearer to the pure mineral than might perhaps be expected from the dark appearance in the hand-specimen. (Ca,Na) is very close to 1 atom; there is a small

TABLE I. Analyses[1] and physical data.

	Pyroxene in calcite.	Pyroxene	Amphibole	Biotite
		from a gneiss-like inclusion.		
Lab. no.	1037	1038	1039	1040
Slide no.	S32453	all from S32451		
SiO_2	54·23	51·04	41·72	39·1
Al_2O_3	1·84	4·74	15·86	17·6
Fe_2O_3	0·92	1·85	3·36	1·1
FeO	1·98	2·79	6·03	6·8
MgO	16·02	14·77	14·14	18·7
CaO	24·02	23·64	12·92	1·2
Na_2O	0·88	0·46	1·42	0·5
K_2O	0·03	0·15	2·60	9·5
$H_2O + 105°$	0·04	0·16	0·85	n.d.
$-105°$	0·05	0·09	0·04	n.d.
TiO_2	0·14	0·39	0·81	1·9
P_2O_5	trace	trace	trace	trace
MnO	0·23	0·17	0·12	0·1
Cl	trace	trace	0·46	0·3
F	trace	trace	0·16	0·2
Li_2O	0·02	trace	0·02	trace
BaO	n.d.	n.d.	n.d.	0·5
	100·40	100·25	100·51	97·5†
Sp. gr. 20°/20°	3·29	3·33	3·175	2·89
α ... ..: ...	1·660*	1·673	1·688	1·659	1·573
β	1·668	1·679	1·694	1·668	—
γ	1·690	1·699	1·711	1·677	1·618
2V	65°	57°+	62°+	86°	—
$Bx_a : c$	40°	38°	45°	22°	—
Axial plane	(010)	(010)	(010)	—
Dispersion	r > v	r > v	slight	—
Colour in section	colourless	faint green	γ bluish-green β yellow-green α yellow	ω brown ε pale straw

* The data in italics were measured on the colourless diopside of the 'white marble', a neighbouring occurrence containing dolomite and forsterite, slide no. S24582. Optical determinations already made on similar but independent material by Sir E. Bailey were in close agreement with the present values throughout, and confirm the representative nature of the specimens; he points out that the data for the white marble diopside indicate the presence of $MgSiO_3$. The biotite is too much crushed to give a reliable axial angle.

† Analysis no. 1040 was carried out on a semi-micro scale and a reliable figure for water could not be obtained, owing to shortage of material.

replacement of Ca by Na, but no enstatite. A small content of Al is explained by the replacement $Al_2/SiMg$. The molecular ratio MgO : FeO exceeds 9 : 1, so that according to H. H. Hess[2] the mineral will be classed as *diopside*.

[1] Published in Summary of Progress for 1938 (Mem. Geol. Surv.), 1939, p. 94.

[2] H. H. Hess, Amer. Min., 1941, vol. 26, pp. 515–535 and 573–594. [M.A. 8–233.]

TABLE II. Analyses calculated to atomic ratios.

	Pyroxene.	Pyroxene.	Amphibole.	Biotite.
Lab. no.	1037	1038	1039	1040
Si	1·966 ⎱ 2·000	1·867 ⎱ 2·000	6·14 ⎱ 8·00	2·78 ⎱ 4·00
Al	0·034 ⎰	0·133 ⎰	1·86 ⎰	1·22 ⎰
Al	0·045 ⎫	0·071 ⎫	0·89 ⎫	0·26 ⎫
Ti	0·004	0·011	0·09	0·10
Fe'''	0·025	0·051	0·37	0·06
Li	0·003 ⎬ 1·009	— ⎬ 1·029	0·01 ⎬ 5·21	— ⎬ 2·80
Mg	0·865	0·805	3·10	1·98
Fe''	0·060	0·086	0·74	0·40
Mn	0·007 ⎭	0·005 ⎭	0·01 ⎭	— ⎭
Na	0·062 ⎫	0·033 ⎫	0·40 ⎫	0·07 ⎫
Ca	0·932 ⎬ 0·995	0·926 ⎬ 0·966	2·04 ⎬ 2·93	0·09 ⎬ 1·02
K	0·001 ⎭	0·007 ⎭	0·49 ⎭	0·86 ⎭
O	5·990 ⎱ 6·000	5·961 ⎱ 6·000	22·98 ⎫	11·00
OH	0·010 ⎰	0·039 ⎰	0·83 ⎬ 24·00	H_2O, &c.,
F	—	—	0·07	omitted
Cl	—	—	0·12 ⎭	
Vol. of cell containing[1] ...	6O = 66·2	6O = 66·0	24O = 279	(12O) = 148

Fassaite of the aggregate (Lab. no. 1038).

Here the proportion of Al is substantial. (Ca,Na) is still close to 1; there is, however, a deficiency of 0·034 which with the excess of 0·029 in the Mg group would imply the presence of a little enstatite. The ratio MgO:FeO is just over 9:1, so that the mineral would still be a diopside as defined by Hess. This classification, however, was devised mainly for igneous pyroxenes, and does not take into account the alumina content, which is probably the most important characteristic for the present minerals. C. E. Tilley[2] has given determinations of two aluminous pyroxenes from Ceylon and Monzoni, and proposes to employ the name fassaite for aluminous pyroxenes with a full content of Ca. These form a numerous and distinctive class. Accordingly, the present mineral may be placed as a member of the fassaite-diopside series (Al/Si 0·133 atom, O = 6) with an important proportion of *fassaite*, while the other pyroxene, described above, belongs to the same series, but is nearly pure diopside.

The optical data for the Monzoni fassaite were α 1·686 (1·688), β 1·694 (1·694), γ 1·712 (1·711), $\gamma:c = 43°$ (45°), sp. gr. 3·319 (3·33), Al_2O_3 % 7·61 (4·74). The physical values in brackets, for the present mineral, agree closely, but these data only confirm the opinion expressed by

[1] See A. F. Hallimond, Amer. Min., 1943, vol. 28, p. 77. [M.A. 9–270.]
[2] C. E. Tilley, Geol. Mag. London, 1938, vol. 75, pp. 81–86. [M.A. 7–181.]

Tilley: 'It is impossible to predict from physical data alone the tenor of alumina in hornfels pyroxenes.' The increase in refractive index over that of pure diopside is no doubt much influenced by the relatively small iron content. The Monzoni assemblage is pyroxene–spinel–calcite, and the present example, with a lower content of alumina, occurs in an assemblage of 'metasilicate' type, without either spinel or free silica.

The two Tiree pyroxenes bear some resemblance to those recorded by H. Eckermann from Mansjö.[1] Of the latter, that from 10 cm. within the limestone is richer in enstatite than no. 1037, and that next the igneous material higher in iron than no. 1038. The variation in MgO/FeO is, however, in the same sense at both localities, the limestone mineral being the more magnesian.

Another similar analysis is given by C. F. Davidson[2] for a diopside from a calcitic paragneiss at Rodil, South Harris. This is shown by Davidson to be similar to the *limestone* diopside from Tiree (not that from the 'xenolith'). It is associated with a pargasite which, apart from an unusually high content of CaO, generally resembles that from Tiree. None of the Harris minerals, however, contains as much iron and alumina as the corresponding ones from the Tiree gneiss-like aggregate, and the refractive indices and density fully confirm this disparity. A pargasite was also obtained from relatively pure limestone, and the two Harris amphiboles reflect the behaviour of the Tiree pyroxenes in that the one derived from the limestone has a lower content of iron and alumina. Davidson regards the paragneiss as a metamorphosed impure limestone, the sodium being possibly derived from a sodic clay constituent.

Pargasite of the aggregate (Lab. no. 1039).

A list of selected calciferous amphiboles has been published by the present writer,[3] and the representative point for the Tiree mineral was plotted as no. 147 in fig. 1 of that paper. The present analysis shows almost exactly two atoms of Ca, and the alkali, in which K slightly predominates over Na, fills the 'vacant space' but does not replace Ca. Silicon has been replaced by aluminium almost to the normal limit at 6Si, so that the mineral approaches the composition of hastingsite and

---

[1] H. von Eckermann, Geol. För. Förh. Stockholm, 1922, vol. 44, p. 354. [M.A. 1–396.]

[2] C. F. Davidson, Trans. Roy. Soc. Edinburgh, 1943, vol. 61, pp. 71–112. [M.A. 9–166.]

[3] A. F. Hallimond, Amer. Min., 1943, vol. 28, pp. 65–89. [M.A. 9–270.]

should perhaps strictly be so termed, though the mode of origin in limestone makes pargasite more convenient. A. N. Winchell[1] and N. Sundius[2] have proposed variations in the detailed nomenclature of the amphiboles. Limestone alkali-amphiboles hitherto often termed pargasite form, however, a very distinctive group (cf. figs. 1 and 2 above mentioned), and the rigorous application of such a nomenclature would set up an arbitrary division of this group with the result that the intermediate (and commonest) members could only be named with certainty with the help of a chemical analysis. For general purposes, therefore, it may be permissible to continue the use of *pargasite* for limestone amphiboles of the present class, even when the content of Al approaches that of hastingsite.

By selecting the points adjacent to no. 147 in the above-mentioned fig. 1, it is possible to collect a group of similar analyses, from which the limestone occurrences may be selected as more directly comparable. No. 152, from Mansjö (Eckermann, 1922, loc. cit.), occurs sparingly near the extremities of reacted pegmatite dikes in limestone, with scapolite, pyroxene, &c. The Mansjö pegmatite contains a good deal of iron, yet the amphibole is highly magnesian. The Mansjö pyroxenes already mentioned show a similar increase in Mg toward the limestone. Yet the limestone itself contains only 1 % MgO. The explanation would seem to be that the magnesia of the original limestone has been almost entirely taken up by the silicates, according to a typical dedolomitization reaction, leaving a calcite-rock, as will be suggested also for Tiree. No. 158, from Pargas (Laitakari, 1921, loc. cit.), is closely similar to Tiree, but, like most Pargas material, is rich in fluorine. No. 156 is from a Monte Somma bomb and no doubt represents the reaction of a limestone fragment with the lava; the analysis is incomplete.

Chlorine has rarely been recorded in an amphibole. Its presence was confirmed by a separate estimation on a sample which had not been cleaned with HCl. An outstanding example seems to be the mineral dashkesanite[3] with up to 7·24 % Cl. (F,Cl,OH) is only half the value required by the usual cell-formula, but this seems to be common in amphiboles. Here, as in some other cases, the difference cannot be attributed solely to oxidation of the iron.

---

[1] A. N. Winchell, Amer. Min., 1945, vol. 30, p. 27. [M.A. 9–271.]

[2] N. Sundius, Sveriges Geol. Undersök., 1946, Årsbok 40, no. 4. [M.A. 10–70.]

[3] G. A. Krutov, Bull. Acad. Sci. URSS, Cl. Sci. Mat. Nat., Sér. Géol., 1936, pp. 341–371. [M.A. 6–438.]

Biotite of the aggregate (Lab. no. 1040).

The present writer[1] has published a diagram for 119 biotites showing the values of RO and R_2O_3 for $(Si,Ti)O_2 = 600$. The diagram was prepared a few years before the micas were investigated by X-ray methods, but the unit cells do in fact commonly contain about $6(Si,Ti)$. In any case it is a rigorous statement of the oxide ratios, and can be used at once to test the agreement of the published analyses with any proposed formulae. The component formulae required by current structural theory are those generally accepted for muscovite and phlogopite, together with a substituted phlogopite $K_2R_5''R_4'''Si_5O_{20}(OH)_4$. The last was termed eastonite by A. N. Winchell[2] who, however, calculated the iron as FeO. Eastonite is represented on the above diagram by the point R_2O_3 240, RO 600, and if a triangle is completed between this point and those representing muscovite and phlogopite it will be seen that the plotted analyses are quite well grouped within this area, so that, as far as regards the oxides plotted, the deviations in composition from simple mixtures of phlogopite and muscovite are adequately expressed by introducing to a small extent the well-known substitution $Al_2/MgSi$.

On this diagram the co-ordinates of the Tiree mica will be R_2O_3 160, RO 497. The point lies about half-way between phlogopite and the most acid biotites, so that in composition the mineral must evidently be classed with the biotites. It is indeed practically uniaxial, but, as S. B. Hendricks[3] has shown, these micas show complex differences in the arrangement of the layer structure, and these will no doubt involve optical variations which will render it impossible to establish a classification of biotite and phlogopite by means of the optic axial angle alone. Neighbouring analyses in the diagram are mainly iron-bearing biotites from veins, gneiss, &c., yet there is here a very high ratio of magnesia. The exceptional nature of the Tiree mineral is fully confirmed by comparison with a recent review by E. W. Heinrich.[4] That author does not discuss the relationship with alumina, which is dealt with in the above diagram, but he has prepared a series of triangles with FeO, MgO, and $Fe_2O_3+TiO_2$ as co-ordinates, on which are represented the micas derived from various rock-types. The present mica will be represented by a point near the MgO corner of the diagrams and it is at once evident that only two types of rock have yielded such micas: the 'ultrabasic'

[1] A. F. Hallimond, Min. Mag., 1926, vol. 21, p. 29.
[2] A. N. Winchell, Amer. Min., 1935, vol. 20, pp. 773–779. [M.A. 6–294.]
[3] S. B. Hendricks, ibid., 1939, vol. 24, p. 729. [M.A. 7–496.]
[4] E. W. Heinrich, Amer. Journ. Sci., 1946, vol. 244, pp. 836–848. [M.A. 10–73.]

and 'limestone' classes. All the ordinary igneous and metamorphic sources yield micas with a considerably higher content of FeO. The Tiree mica may therefore be classed as an abnormally magnesian *biotite*.

IV. EQUILIBRIUM RELATIONS.

Analyses of amphiboles accompanied by that of the associated pyroxene or mica are not common. A few examples bearing on the present question are given in table III.

TABLE III. Partition of oxides between amphibole (Am.), pyroxene (Pyr.), and mica (Bi.).

Oxide. Wt. %.	Tiree (147).			Mansjö (152).			Edenville (96).		Vesuvius (156).		Khibina.[1]	
	Pyr.	Am.	Bi.	Pyr.	Am.	Bi.	Pyr.	Am.	Pyr.	Am.	Pyr.	Am.
MgO ...	14·8	14·1	18·7	17·0	17·8	26·6	10·0	11·5	13·7	16·5	14·1	14·9
FeO ...	2·8	6·0	6·8	4·1	3·2	0·6	12·8	13·8	—	—	1·9	1·6
Fe$_2$O$_3$ ...	1·8	3·4	1·1	0·5	1·5	2·4	1·3	3·1	—	—	8·7	7·1
Al$_2$O$_3$ ...	4·7	15·9	17·6	1·6	15·4	14·7	2·0	11·9	5·6	8·3	3·4	16·3
CaO ...	23·6	12·9	1·2	22·6	13·8	0·43	22·1	11·6	22·9	14·5	22·4	9·3

The numbers in brackets refer to the list of amphiboles (A. F. Hallimond, 1943, loc. cit.).

The Edenville amphibole is less aluminous than the others, but shows a similar partition with the pyroxene. The monchiquite minerals from Khibina tundra have been included for comparison, but the ratio Fe$_2$O$_3$:FeO indicates that the amphibole is basaltic. The high oxidation is reflected in the pyroxene. E. S. Larsen and others[2] have noted a similar high proportion of Fe$_2$O$_3$ in biotite occurring with basaltic amphibole. The Mansjö pyroxene cited was that in limestone 10 cm. from contact, not occurring directly with the amphibole and mica. The other Mansjö pyroxene is chemically much nearer to the pegmatite with which it is in direct contact.

The first three examples are typical. Their oxide ratios are in many ways consistent and seem in keeping with the view that the associated minerals were in each case approximately in equilibrium. In Khibina the low values for FeO are deceptive, being due to the oxidizing conditions, and the Mansjö mica may also have been subjected to some oxidation, for an associated phlogopite (no. V) is described as 'gold-glanzende'. Alumina enters the amphibole and mica much more freely than the pyroxene, while the other oxides appear to be fairly evenly

[1] N. P. Lupanova, Trav. Inst. Pétrogr. Acad. Sci. URSS, 1934, no. 6, pp. 53–64. [M.A. 6–420.] Phenocrysts in a monchiquite dike.

[2] E. S. Larsen, I. Irving, F. A. Gonyer, and E. S. Larsen III, Amer. Min., 1937, vol. 22, p. 895. [M.A. 7–31.]

shared, with rather more MgO in the mica. The CaO approximates to saturation in pyroxene and amphibole, and is, of course, practically absent in the mica.

V. GENERAL CONCLUSIONS.

The Tiree diopside contains so little alumina that the groundmass marble in which it occurs must be regarded as substantially similar to the 'white marble' (cf. Coomáraswámy, 1903, loc. cit.), though apparently without any local excess of magnesium capable of yielding orthosilicate minerals. The only important foreign constituent indicated seems to be fairly pure silica, or siliceous inclusions, scattered in a remarkably even manner throughout the rock.

In the gneiss-like aggregate the abnormally high magnesia points to a free reaction with the limestone, which has been dedolomitized. Evidently, however, the other oxides, particularly the alumina, must here be derived almost entirely from the original rock-fragment which was metamorphosed to form the aggregate. The amphibole is fully alkaline and there is a further amount of potassium in the mica, so that the quantity of alkali would agree with an orthogneiss (or its unaltered equivalent) as a possible original material of the inclusion. On the other hand, the possibility of derivation of the aggregates from an alkaline sediment such as that of Harris (C. F. Davidson, 1943, loc. cit.) may require consideration. When allowance is made for the changes due to crushing, the introduction of chalcedony, and the alteration of the scapolite, the marble seems to be essentially a metamorphic limestone of an intermediate grade. During the metamorphism a free chemical readjustment took place, not only by transfer of MgO and of $CaCO_3$ to the silicates by dedolomitization with scapolitization, but also by a local movement of the 'volatiles' leading to alternative growth or recession of pargasite, mica, and fassaite. These changes seem to be proceeding simultaneously in different places even in the same rock section. The reaction is particularly characterized by extended intergrowths of pargasite and mica, which tend to obliterate the normal equilibrium 'pflaster' structure. Whether the latter was already present in the original fragment or was only developed during the last metamorphism may be difficult to decide. The present evidence seems clear, on chemical as well as mineralogical grounds, that a general metamorphism accompanied the dedolomitization, leaving the silicate minerals in local chemical and textural equilibrium with the groundmass of coarse calcite.

In the distribution of the oxides between minerals and limestone, and texturally, as, for example, in the intergrowth of mica and pargasite, there is a strong resemblance to the Mansjö occurrence. At that locality various highly magnesian silicates also occur at the contact of an iron-bearing acid igneous rock and a calcite-marble. The close analogy with the Tiree marble would seem to indicate that the Mansjö minerals have gained their magnesium from the limestone by a metamorphic reaction after emplacement of the pegmatite, rather than by progressive modification of the pegmatite during injection. The series of pegmatite analyses given by Eckermann (1922, loc. cit., p. 326) support the view there expressed that potassium has migrated from the pegmatite and has been precipitated as phlogopite near the contacts. On the other hand, there seems no similar evidence for the migration of the very small amounts of magnesia shown in the analyses: the limestone seems the most probable source of this constituent, which, like the potassium, has been precipitated as insoluble silicates at the contact.

The Mansjö pargasite-mica intergrowths lie in calcite at the centre of thin apophyses of the pegmatite, so that they resemble a late liquid residue. Since the formation of the silicates yields free CO_2, it is always possible that a small development of carbonate liquid may have taken place, with the formation of calcite veins of very local significance. In other cases there may be only a weakening of the limestone at the time of reaction. It has been shown that calcite is more easily deformed under such conditions even when far below the melting-point.

The most remarkable feature of the Tiree marble is the extensive distribution of the foreign rock-fragments. The present investigation seems to establish their emplacement before the latest metamorphism, but little additional information has been gained as to the original mode of emplacement. A small proportion of other rocks such as pegmatite are also present, and this basic aspect of the problem might perhaps be decided by a general survey of the nature and distribution of the 'xenoliths', with allowance for metamorphic changes of the kind indicated in the present paper, operating on what was originally a dolomitic limestone of a type that is widely distributed in the Lewisian rocks.

Note on the dilution of methylene iodide with aniline.

Volatile solvents have commonly been employed to lower the constants of methylene iodide in mixtures used for work on densities and refractive indices. They have the disadvantage of requiring distillation or freezing

to recover the iodide, and an alternative was sought in the course of the work on the Tiree minerals. Aniline as a diluent seems to fulfil all the requirements fairly well. It is only moderately volatile, and can be removed from the methylene iodide by washing the liquid in a separating funnel with dilute hydrochloric acid. This forms water-soluble aniline hydrochloride, while practically pure iodide sinks and can be drawn off. The iodide so obtained contains a little hydrochloric acid, which is removed by washing with weak ammonia, after which the iodide is dried by contact with fused calcium chloride. A sample recovered in this way in 1938 was found to be in good condition after nine years, with normal refractive index, though wine-coloured; it cleared well on standing with a piece of copper wire.

Note.—Since this paper was read, a review of the biotites from igneous rocks has been published by S. R. Nockolds (Amer. Journ. Sci., 1947, vol. 245, pp. 401–420). Only five of the 77 micas there listed have a ratio (total) FeO:MgO as low as that from Tiree, so that the conclusion already drawn is confirmed. Nockolds's diagrams indicate a strong tendency for hornblende (in igneous rocks) to occur only with the less magnesian biotites. The Tiree paragenesis stands in marked contrast with this relation, since hornblende occurs closely intergrown with a highly magnesian biotite. Furthermore, Nockolds indicates a well-marked upper limit to the alumina ratio of the igneous biotites that occur with hornblende, diopside, or olivine; the Tiree mica again seems exceptional in having an alumina ratio somewhat above this limit. These peculiarities tend to confirm the view already expressed that the Tiree aggregate was formed by chemical alteration at a lower temperature, and not directly from fusion, though the original material may well have been a fragment of orthogneiss.

EXPLANATION OF PLATE XIV.

Figs. 1 and 2, Microstructure of groundmass of Tiree marble. Figs. 3–6, Microstructure of aggregate from Tiree marble.

The numbers refer to slides and photographs in the Geological Survey Collection.

FIG. 1. Pyroxene, polyhedral grains with fissures containing calcite and chalcedony. One large grain of strain-twinned calcite. Crossed polarizers. S32453. M2300. ×27.

FIG. 2. Pyroxene, twinned || a, with an embayment of coarse twinned calcite, in crushed calcite. Crossed polarizers. S32453. M2302. ×27.

FIG. 3. Amphibole with envelope of altered scapolite containing grains of pyroxene, in crushed calcite. Small idiomorphic amphibole with a partial envelope, in calcite. Polarized light. S32451. M2299. ×27.

FIG. 4. Pyroxene with dark inclusions, also small grains of amphibole and mica, in partly crushed calcite. Polarized light. S32451. M2298. ×27.

FIG. 5. Polyhedral aggregate of altered scapolite (top), amphibole (middle), two rounded grains of calcite (middle, right), pyroxene (bottom, dark), with strained calcite (bottom, left). Crossed polarizers. S32451. M2297. ×27.

FIG. 6. Parallel intergrowth of mica and amphibole, with altered scapolite. Crossed polarizers. S32451. M2301. ×27.

Plate XIV.
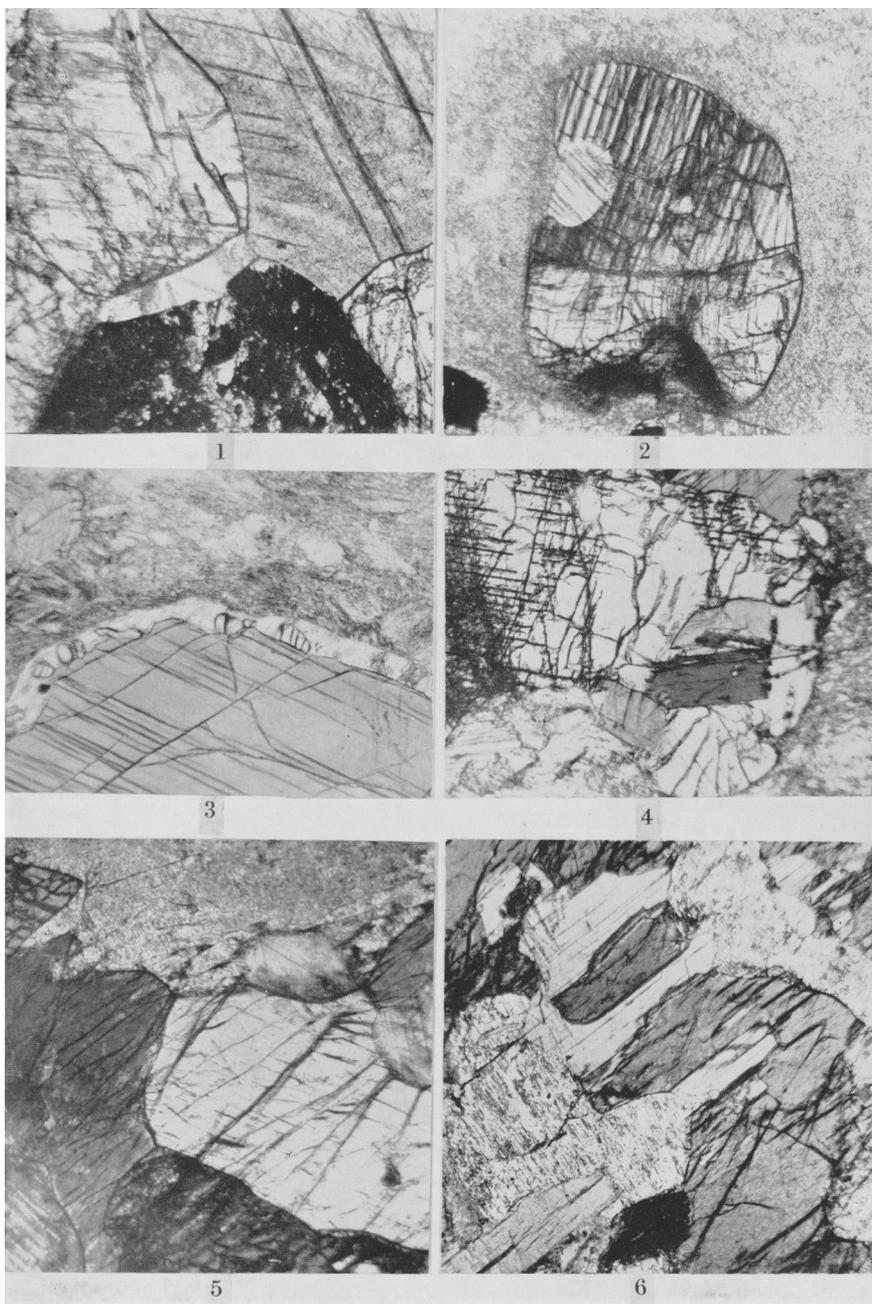
A. F. HALLIMOND: PYROXENE, ETC., IN TIREE MARBLE.