Mineralogy and petrology of the Highland Border Suite serpentinites

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ABSTRACT. Prior to serpentinization, the ultramafic rocks of the ophiolitic Highland Border Suite were depleted harzburgites, dunites, and orthopyroxenites. They may have been the residual mantle after melting of primary lherzolite that produced the basaltic rocks of the Suite. Serpentinization of these peridotites occurred at low temperatures (75 ± 50 °C) and produced pseudo-morphic-textured lizardite serpentinites in which the minor-element compositions of the primary phases were preserved in the lizardite grains. Olivine was almost completely serpentinized before orthopyroxene was hydrated. Concomitant peripheral alteration of chrome-spinels, during serpentinization of orthopyroxene, caused release of Cr and Al cations which were incorporated into the lizardite. Under greenschist-facies conditions (c. 350 °C at 3-4 kbar), the lizardite serpentinites were subsequently metamorphosed and recrystallized to non-pseudomorphic-textured antigorite serpentinites. Post-metamorphic processes included high-level brittle deformation that resulted in fracturing and cross-fibre chrysotile vein-formation, and ductile shearing ($T \sim 200$ °C, $P > 2$ kbar) which produced talc-chrysotile shear zones.

THE mafic and ultramafic rocks that outcrop along the Highland Border Fracture Zone (H.B.F.Z.)—The Highland Border Suite (Ikin, 1979)—have been interpreted as ophiolitic remnants of a Caledonian marginal basin of Cambrian or early Ordovician age (Henderson and Robertson, 1982; Curry et al., 1982; Ikin, 1983). Rocks of the Suite occur as tectonically emplaced fault-bounded slivers and pods varying in width from 30-500 m, and separated one from another by as much as 35 km. The H.B.F.Z. is a complex zone of faulting, monoclinal downwarping and unconformity which marks the boundary between the late Precambrian–Cambrian Dalradian metasediments of the exposed ortho-

tectonic Caledonides to the north, and the gently folded Upper Palaeozoic sedimentary basin of the Midland Valley to the south (fig. 1).

The presence of ultramafic rocks along the H.B.F.Z. was first recognized over 150 years ago (Lyell, 1825). During the following 100 years, geologists (and particularly those working for the Geological Survey) provided petrographic descriptions of these rocks from a number of different localities, e.g. Kirriemuir (Judd, 1885), Aberfoyle (Jehu and Campbell, 1917), and Bute (Smellie, 1916). However, subsequent studies focused on the structural relationships between the Highland Border Suite (H.B.S.) and associated sediments, and the adjacent Dalradian and Old Red Sandstone rocks. In consequence, little new mineralogical or petrological data on the ultramafic (and mafic) rocks were collected. Nevertheless, it was during
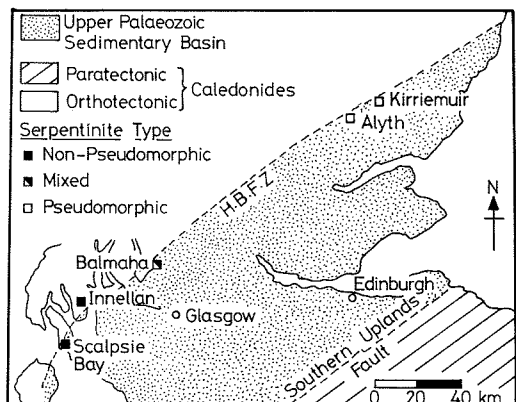

FIG. 1. Regional map of central Scotland showing major geological features, position of five localities discussed in this paper, and major cities.

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this period that Allan (1928) and Anderson (1947) first proposed that the separate outcrops of ultramafic rock along the H.B.F.Z. were all 'related' to each other and, hence, originally part of a formerly more extensive igneous body.

In the 1970s, interest in the Highland Border rocks was renewed because of their possible ophiolitic status, and a number of tectonic models of the Caledonides were constructed in which the Highland Border rocks were interpreted as remnants of an ocean, or marginal, basin (e.g. Bird et al., 1971; Garson and Plant, 1973). Such models, however, were proposed, to a large extent, merely on the existence of an assemblage of serpentinites, spilites, cherts, etc., and, most significantly, in the absence of detailed geochemical, mineralogical, and petrological data.

Recently, however, this situation has been partially rectified by new investigations of the Highland Border rocks; these have included stable-isotope (Ikin and Harmon, 1981, 1983), petrochemical (Ikin, 1983), and petrological, structural, and sedimentological (Henderson and Robertson, 1982) studies. As a contribution to these investigations, this paper presents new petrological and mineralogical data on some of the H.B.S. ultramafic rocks.

The ultramafic portion of the Highland Border Suite is dominated by lizardite serpentinites some of which have been metamorphosed to antigorite serpentinites, although some rocks have been preserved in which hydration was not total. In this paper, we discuss (i) the ultramafic rocks' primary phase mineralogy as deduced from textural and chemical studies of relict minerals in the partially hydrated serpentinites, and (ii) some chemical aspects of the serpentinization process, and the mineralogy and petrography of the serpentinites. We show that the H.B.S. serpentinites were derived from chemically depleted peridotites and that during their evolution they occupied several different crustal levels (and hence P-T conditions), being subjected to near-surface hydration (serpentinization), mineralogical, and textural reconstitution at depth (metamorphism) and subsequent re-equilibration at higher crustal levels during later, but varying forms of, deformation.

General characteristics. The H.B.S. serpentinites comprise both the pseudomorphic and non-pseudomorphic types of Wicks and Whittaker (1977). The former, in which serpentine occurs as a direct pseudomorph of the primary silicates, are found at Alyth and Kirriemuir (fig. 1). The latter, in which the serpentinite fabric is unrelated to the primary textures in the ultramafic precursor, are found at Innellan and Scalpsie Bay, Isle of Bute. Important evidence concerning the relationship

between the two different fabric-types occurs at Balmaha, where 'mixed-type' serpentinites contain varying amounts of randomly orientated (non-pseudomorphic) blades of serpentine that grow across, and at the expense of, an underlying pseudomorphic fabric.

In the pseudomorphic serpentinites, relict primary phases have been preserved, and the pseudomorphic replacement by serpentine of primary igneous textures has resulted in a high degree of preservation of original grain-boundary relationships. Thus, primary rock-types can be distinguished. Harzburgites were dominant, making up at least 90% of all the H.B.S. ultramafic rocks examined, with the remaining 10% consisting of dunites and orthopyroxenites (rock nomenclature in accordance with Jackson's (1968) classification scheme). In the harzburgites, modal orthopyroxene varied between 8 and 30%, clinopyroxene and chrome-spinel being present in only accessory ($\leqslant 2\%$) amounts. Textures were generally xenoblastic granular with olivine and orthopyroxene exhibiting interlocking grain boundaries, and cuspate boundaries and embayments being indicative of limited grain-boundary reaction.

A significant proportion of the harzburgites, however, had a porphyroclastic texture in which large strained orthopyroxene grains occurred in a finer- and equigranular-grained assemblage of interlocking olivine, orthopyroxene, and spinel. Many of the relict orthopyroxene porphyroclasts are strained, showing undulose extinction and fractures, along which recrystallization has taken place. These porphyroclasts often contain exsolution lamellae of clinopyroxene and the remnants of narrow (< 1 mm) clinopyroxene reaction rims. There is a sub-parallel orientation of the large orthopyroxene grains in some parts of the mass at Alyth and Carity Burn, Kirriemuir. However, because of their porphyroclastic nature, we are not sure whether this is an igneous phenomenon (crystal settling?) or the result of tectonism, although the occurrence of some mineralogical layering (i.e. variations in olivine/orthopyroxene ratios) at Carity Burn would lend some support to the 'crystal settling' interpretation.

Mosaic/granuloblastic-textured dunite occurs as layers (5–20 cm thick) that are sub-parallel to the layering described above. Because of the high degree of serpentinization it is impossible to say whether they formed from extreme compositional layering or as concordant intrusive sill-like bodies. Orthopyroxenites at Alyth occur as pod-like segregations, but at Balmaha they form discordant dykelets or veins; at Carity Burn (Kirriemuir), both types have been observed, the latter running at, or near, right-angles to the crude layering.

In summary, the harzburgites are characterized by a rather patchy distribution of orthopyroxene and some evidence of deformation and recrystallization. The textures and mineralogy are similar to those found in peridotite bodies which were formerly described as 'Alpine-type', and now considered to be the ultramafic components of dismembered ophiolite complexes.

Primary phase chemistry. Although nearly all the pseudomorphic serpentinites contain relict spinels, out of approximately sixty samples examined, only one contained relict olivine and eight relict orthopyroxenes. Representative electron microprobe analyses of these phases, in harzburgites from Alyth, are presented in Table I. The analyses were performed on a (wavelength-dispersive) Cambridge Instruments Microscan V microprobe at the University of Leicester, and corrections to raw data were made using a MAGIC IV programme (Colby, 1968).

In spite of the high degree of serpentinization of the rocks in which these minerals occur, we believe that the analyses in Table I reflect the original igneous mineral chemistries, and have been unaffected by 'leaching' caused by the circulating fluids. First, the figures in Table I are mean compositions, but have been derived from a number of 'point analyses' from a number of grains of each phase in each sample. We have observed, however, that mineral compositions within each sample are remarkably constant with inter-grain and within-grain compositional variations being insignificant. If leaching of the primary phases by the circulating fluids of serpentinization had occurred, we would have expected a much greater variation in chemistry both within and between grains. Secondly, and related to the first point, there is little variation in composition between 'point analyses' in the centre of relict grains and those at the margins of grains immediately adjacent to the serpentine. It is in the latter areas that we would have expected any significant alteration (leaching) to occur.

This apparent resistance of the primary phases to circulating fluids, however, is not surprising. Ikin and Harmon (1981, 1983) have shown that there was probably little, or no, O-isotope exchange between the ultramafic rocks as a whole, and the fluids of serpentinization. Additionally, serpentinization probably occurred at a low temperature ($\leqslant 150\,°C$), which would have made selective leaching less likely.

TABLE I. *Representative electron probe analyses of primary phases in serpentinized harzburgites from Alyth*

| | A312 | | | | A302 | | A2 | |
| | Olivine | | | | | | | |
	Core	Rim	Orthopyroxene	Spinel	Orthopyroxene	Spinel	Orthopyroxene	Spinel
SiO_2	41.21	41.13	57.58	0.10	57.51	0.07	57.06	0.04
Al_2O_3	0.01	0.02	0.80	16.80	1.37	24.90	1.19	17.04
Cr_2O_3	n.a.	n.a.	0.33	51.17	0.46	43.31	0.51	52.28
FeO*	8.31	8.26	5.54	18.60	5.31	18.54	5.18	17.48
MnO	0.11	0.11	0.12	0.52	0.12	0.44	0.08	0.48
MgO	49.25	49.86	33.96	10.09	33.65	11.05	34.70	10.81
CaO	0.03	0.05	0.70	0.02	0.68	0.03	0.95	0.03
NiO	0.22	0.19	0.04	< 0.01	< 0.01	< 0.01	0.01	0.03
Total	99.14	99.62	99.07	97.31	99.11	98.35	99.68	98.17
Fo (%)	91.4	91.6	—	—	—	—	—	—
100 Cr/(Cr + Al)			21	67	18	54	22	67
100 Mg/(Mg + Fe^{2+})			—	50.5	—	52.4	—	53.3
Wo (%)	—	—	1.3	—	1.3	—	1.8	—
En	—	—	90.5	—	90.7	—	90.6	—
Fs	—	—	8.2	—	8.0	—	7.6	—

n.a. not analysed.
* Total Fe as FeO.
Operating conditions: accelerating voltage 15 kV, specimen current $\sim$ 20 nA, counting time 30 sec.
Natural and synthetic standards used for calibration.

The composition of olivine, in the one sample in which it was observed (A312), is in the range $Fo_{91.2-91.7}$. Inter-grain variation is in the order of 0.2% Fo, with no significant variation in core-rim compositions. Thus, we consider the olivine grains to be homogeneous and of uniform composition. The same is true of the orthopyroxenes in each of the three samples analysed. The major cation content varies little, with all grains falling in the range $Wo_{0.8-2.6}En_{89.7-91.3}Fs_{7.5-9.2}$. The main difference between the orthopyroxenes from the three samples is in their minor-element contents, Al in particular and Cr to a lesser extent. The spinels have magnetite rims that formed during serpentinization; thus, only core rims have been analysed. The Mg/Fe ratios in each of the three samples are almost identical; however, although samples A312 and A2 have identical Cr and Al contents (and hence $Cr/(Cr+Al)$ ratios), sample A302 is different. Despite this, the $Cr/(Cr+Al)$ ratios of the three samples (0.54 to 0.67) are within the (0.45 to 0.85) range for spinels from altered and unaltered serpentinites and serpentinite conglomerates from Balmaha (Henderson, Fortey and others, in press). This lends some support to the long held view that the fragmented outcrops of ultramafic rocks along the H.B.F.Z. are the dismembered remnants of a large, and formerly more extensive, igneous complex.

The chemistry of the spinels and orthopyroxenes, and the position that they occupy in the 'dis-crimination diagrams' in figs. 2 and 3, therefore, support the textural and mineralogical evidence, which favours an origin in an 'Alpine-type' ultramafic body (see above for modern interpretation of this term). The low Al content of the orthopyroxenes and the low modal abundance of primary aluminous and calcic phases in these ultramafic rocks, is indicative of a highly depleted bulk-rock composition. Using the known modal mineral contents in the three samples, we have estimated that their total Al_2O_3 content varies between only 0.37 and 0.47%. When these values are compared with the Al_2O_3 content of 'undepleted' primary lherzolite and pyrolite—3.5 to 5.8% (Ringwood, 1975)—it is clear that the H.B.S. ultramafics are of a highly refractory nature. It is possible that these depleted ultramafics represent the residue after partial melting of a primary pyrolite composition that produced the basaltic rocks of the Highland Border Suite.

Geothermometric estimates based on olivine-spinel Mg-Fe distribution indicate an equilibration temperature of $1124\pm36\,^\circ C$ using the thermochemical data given by Jackson (1969), or $644\pm18\,^\circ C$ using the more-recently calibrated thermochemical data of Roeder et al. (1979). The latter, lower, temperature would probably reflect sub-solidus exchange, a feature experimentally verified by the authors. Estimates of equilibration pressures are more difficult to obtain. The method most commonly used is based on the solubility of Al
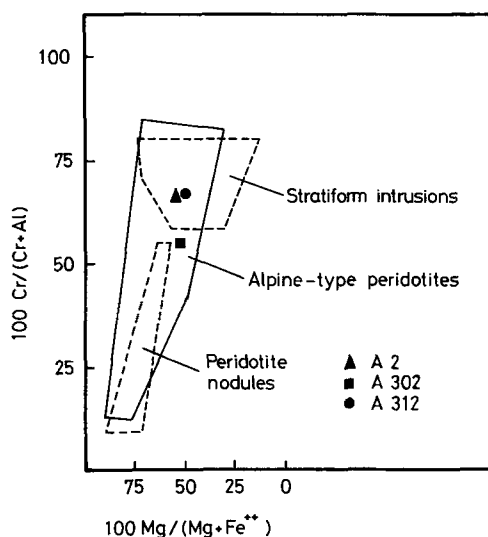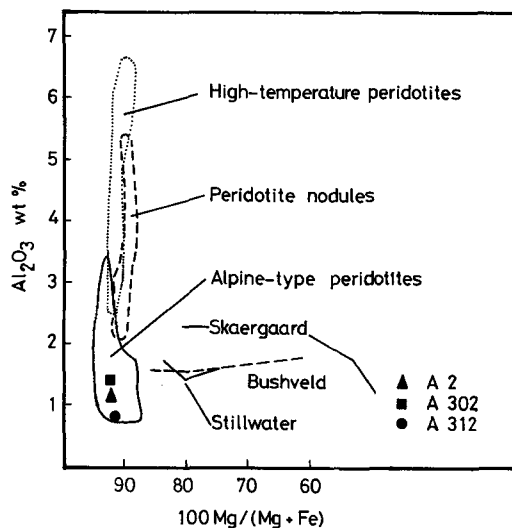
FIGS. 2 and 3. FIG. 2 (*left*). Relationship between mafic index and Al content of orthopyroxenes from harzburgites at Alyth. Points for each sample are mean analyses from a number of grains. Fields from Medaris (1972). FIG. 3 (*right*). Relationship between mafic index and $Cr/(Cr+Al)$ ratio in spinels from harzburgites at Alyth. Points for each sample are mean analyses from a number of grains. Fields from Irvine and Findlay (1972).

in pyroxenes in the presence of an additional aluminous phase. However, Sinton (1977) has argued that the partitioning of Al between pyroxenes and spinel is not solely pressure-controlled, but may be influenced by bulk-rock composition and temperature. Furthermore, pressure estimates have to be made using an independently-derived equilibration temperature value, and in view of the discrepancy resulting from different sets of thermochemical data (see above) there must be some doubt as to whether this can be achieved. In view of these uncertainties, a precise pressure estimate for the H.B.S. peridotites is not made, but some limits can be placed on its value. All the H.B.S. rocks are spinel-bearing, and no garnet-, or plagioclase-bearing types have been found. Using O'Hara's (1967) pyroxene grid, it is, therefore, reasonable to assume that these rocks formed (or equilibrated) at pressures of between 9 and 19 kbar (at $T = 1124\,°C$) or 8 and 12 kbar (at $T = 644\,°C$).

Serpentinization. Serpentinization has produced mesh-texture serpentine from olivine, and bastite serpentine from pyroxene. Concurrently, magnetite rims formed on Cr-spinel grains, and fine disseminated magnetite was 'deposited' along mesh rims and bastitized-pyroxene cleavage planes. The mineralogy of the serpentinites has been investigated by X-ray diffraction techniques (Ikin, 1979; Ikin and Harmon, 1981). All samples and three standards (antigorite, lizardite, and chrysotile) were examined over the range 10–62° 2θ (Cu-Kα radiation) where three segments (18–21°, 34–38°, 58–62°) proved to be the most useful in discriminating between the different serpentine-group minerals.

It is clear from fig. 4 that the pseudomorphic serpentinites are composed almost exclusively of lizardite. In sample CB106, serpentine from mesh-texture (CB106M) and bastite (CB106B) areas was extracted separately. The basic pattern is the same in both but they differ in that CB106M appears to contain a very small amount of brucite. The absence of this phase in the bastite lizardite probably resulted from the orthopyroxene having a higher silica content which inhibited brucite growth. The marginally greater amount of magnetite produced in the mesh-texture areas (fig. 4) may have been due to differential cation liberation between olivine and orthopyroxene since the former liberates a significant proportion of its cation content during topotactic serpentinization, whereas the latter does not (Wicks and Whittaker, 1977).

Lizardite-type serpentinization is commonly believed to occur at temperatures between 25 and 200 °C (Moody, 1976; Wenner and Taylor, 1971). This also seems to be true of the H.B.S. serpentinites, where stable-isotope evidence indicates that
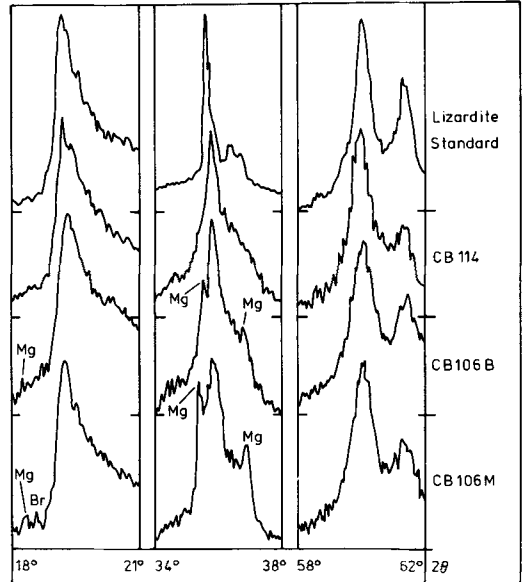

FIG. 4. X-ray diffraction patterns of representative samples of pseudomorphic serpentinites from Carity Burn, Kirriemuir, and the pure lizardite standard. Reflections of accessory minerals are labelled; Br—brucite, and Mg—magnetite.

serpentinization occurred through the interaction of peridotites with waters of meteoric origin at temperatures of $75 \pm 50\,°C$ (Ikin and Harmon, 1981, 1983).

Lizardites from mesh-texture and bastite positions have been analysed in each of the three samples for which chemical data on the relict igneous phases has been presented. Representative electron microprobe analyses of these lizardites are presented in Table II. The lizardites produced from both the olivine (mesh) and orthopyroxene (bastite) are broadly similar in terms of their major cation chemistry, i.e. Mg, Fe, and Si. However, in each of the two textural positions, the serpentine grains retain the minor element 'fingerprints' of their precursor, and this is particularly true of Cr and Al whereby the bastite lizardite is noticeably enriched in both these elements compared with the mesh lizardite. This feature of retaining the minor-element differences between the primary phases during serpentinization, is further enhanced by a selective Cr- and Al-enrichment in the bastite-lizardite (cf. Tables I and II). We attribute this to the release of these cations during the peripheral breakdown of Cr-spinel (to magnetite) during serpentinization and note that of the three samples, the one showing least total (Cr + Al) enrichment in the bastite lizardite (A2) also has the lowest modal

TABLE II. *Representative electron probe analyses of lizardites in serpentinized harzburgites from Alyth*

	Mesh-texture		Bastite		
	*(A312, A302, A2)		A312	A302	A2
	Core	Rim			
SiO_2	40.90	40.96	40.06	39.77	40.32
Al_2O_3	0.09	0.09	1.16	2.23	1.37
Cr_2O_3	0.01	0.01	0.53	0.66	0.63
†FeO	4.63	6.41	7.09	6.57	6.75
MnO	0.07	0.09	0.06	0.21	0.06
MgO	40.41	38.74	37.58	37.25	37.49
CaO	0.04	0.03	0.06	0.12	0.05
NiO	0.26	0.14	0.01	< 0.01	0.02
‡H_2O^+	13.59	13.53	13.45	13.19	13.41
$\dfrac{100\ Mg}{(Mg + Fe^{2+})}$	94.1	91.6	—	—	—

* Mean analyses from three samples.
† Total Fe as FeO.
‡ H_2O^+ calculated by difference.
Operating conditions as in Table I except counting times reduced to 10 sec.

spinel content. This suggests that the degree of enrichment is related to the degree of spinel-rim alteration, and hence roughly proportional to modal spinel content. It is, therefore, apparent that Cr and Al cations are free to exchange between adjacent grains of Cr-spinel and bastite lizardite, and are presumably carried as complexes in the circulating fluids causing serpentinization.

In contrast to the bastite lizardites, it is noticeable that the mesh-texture lizardite shows only minor Cr- and Al-enrichment (over olivine). This could be because olivine was more or less completely serpentinized before orthopyroxene was hydrated and the spinel rims altered to magnetite. This behaviour would be consistent with petrographic observations on virtually all types of ultramafic rock, which indicate that orthopyroxenes are altered at a later stage than olivine during serpentinization.

In the mesh-texture serpentine, the mesh rims are iron-rich compared with the mesh cores (Table II). If, as in sample A312, the olivines were unzoned, then this feature cannot be attributed to an inhomogeneity being inherited by the lizardite, and must have been a product of the serpentinization process. The value of 100 Mg/(Mg + Fe) in the mesh-rim lizardite in each of the three samples analysed is equal to, or marginally greater than, the equivalent ratio (Fo content) in the primary olivines from sample A312. This suggests that in the early stages of serpentinization, in which alteration

is initiated at grain boundaries, most of the iron in the olivine was incorporated into the lizardite lattice. In contrast to the mesh rims (100 Mg/(Mg + Fe) $\sim$ 91.6), the mesh cores are noticeably iron-depleted (100 Mg/(Mg + Fe) $\sim$ 94.1) and it was at this stage, when the olivine cores ($Fo_{91.2-91.7}$) were hydrated, that most of the free iron was released, thus forming magnetite or Fe-brucite.

It is unclear why there is this difference in the extent of iron substitution into the lizardite lattice, between the mesh core and mesh rim. However, there are textural differences (e.g. degree of crystallinity, grain size and orientation) between the lizardite in these two sites, with the boundary between the mesh rim and mesh core possibly representing a pause in the serpentinization process. Martin and Fyfe (1970) and Wicks and Whittaker (1977) have suggested that these types of textural change may result from variations in temperature and water availability, and a shift in hydration reaction from being interface- to diffusion-controlled. These factors may also have been responsible for causing a change in the way in which iron was incorporated into the lizardite lattice.

A schematic summary of the inferred chemical processes which took place during serpentinization, is presented in fig. 5.

Metamorphism. The non-pseudomorphic serpentinites from Innellan and Scalpsie Bay, Bute are mainly composed of antigorite (fig. 6), whereas the 'mixed-type' rocks from Balmaha contain a mixture of lizardite and antigorite (fig. 7). In the X-ray diffraction scans, two of the most distinctive antigorite reflections are those of the composite 003, 18.00 line at *c.* 37° 2θ and 24.30 at *c.* 59° 2θ. These lines are marked in the scans for the 'mixed-type' rocks from Balmaha (fig. 7). In each of the three samples, there are differences in the 'peak heights' and 'areas under peaks' corresponding to these two diagnostic antigorite reflections. On a qualitative basis, therefore, this indicates that the amount of antigorite present in each of the three samples is different. In the serpentinites from Innellan, Scalpsie Bay, and Balmaha, we have been able to estimate the proportions of antigorite and lizardite through petrographic (point-counting) examination and quantitative analysis of X-ray data (Ikin, 1979). The antigorite content in these metamorphosed rocks varies between 18 and 100%.

The petrographic and X-ray data, therefore, indicate that the two 'end-member' types of serpentinite differ in both fabric-type and mineralogy. The original serpentinization resulted in the production of pseudomorphic lizardite serpentinites; this was followed by a later phase of metamorphic recrystallization and antigorite growth. At Balmaha, the
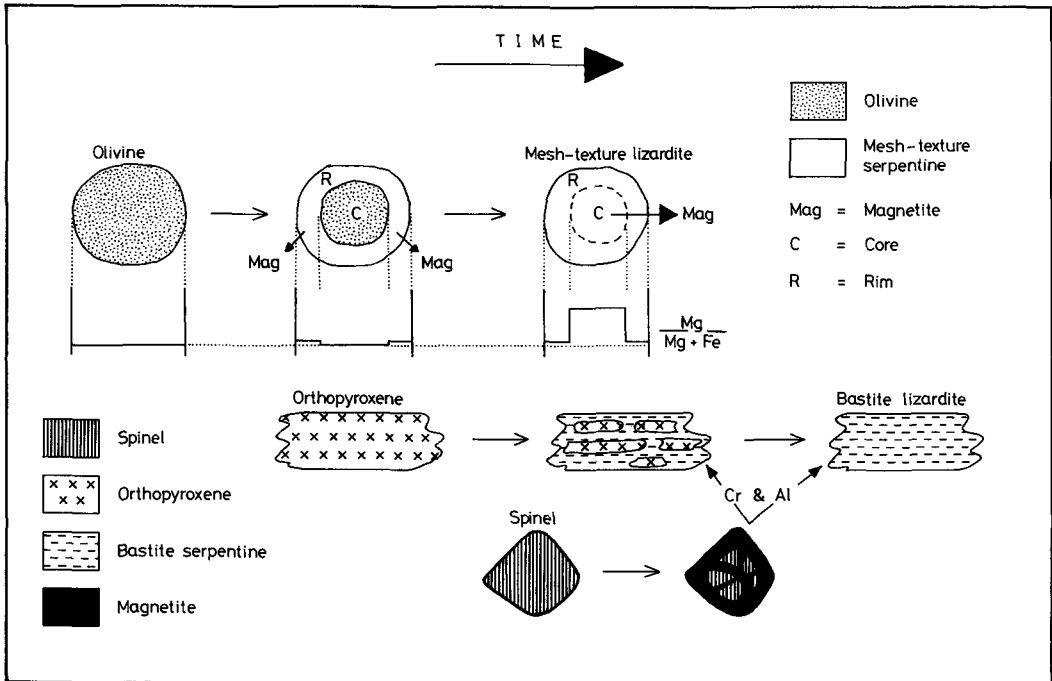
FIG. 5. A schematic illustration of the postulated physical and chemical processes occurring in the primary phases during serpentinization.

serpentinites record only partial metamorphism, and the textural relationship between the pre-metamorphic mineral assemblage and the 'new' metamorphic assemblage is well preserved. However, even in the pure non-pseudomorphic antigorite serpentinites from Scalpsie Bay, there is textural evidence that they were derived from pseudomorphic serpentinites similar to those from Alyth and Carity Burn. Under cross-polarized light, these rocks appear to be a completely randomly orientated interpenetrating mass of antigorite 'blades'; but in plane light, a fine network of disseminated magnetite is seen to delineate a mesh-texture framework. This bears no relationship to the present textural form of the serpentine in these rocks, and must, therefore, reflect an earlier pseudomorphic fabric.

The X-ray diffraction scans (figs. 6 and 7) show that brucite is present in some of these metamorphosed serpentinites. In the least metamorphosed rocks from Balmaha (e.g. BA402), finely interlayered brucite–antigorite plates have been observed replacing lizardite in mesh cores, although no brucite reflections are present in the XRD scans. Such a feature can be explained by the brucite content being close to the mineral's (< 1 %) detection limit (Aumento, 1970). In the 'mixed-type'

rocks that have been more thoroughly recrystallized (e.g. BA3), brucite occurs as discrete grains between interlocking or interpenetrating antigorite blades, and forms a prominent reflection in the XRD scan (fig. 7). However, in the more completely 'antigoritized' rocks (e.g. SBSS1 with 95 % modal antigorite, fig. 6), the petrographic and XRD evidence indicates the presence of little, or no, brucite. Antigorite and brucite appear to increase in quantity together, at the expense of lizardite, up to the point where there is approximately 60 % modal antigorite; thereafter, although antigorite continues to replace lizardite, brucite diminishes in quantity.

Thus, the early and middle stages of recrystallization are described by the reaction:

$$17 \text{ lizardite} \rightleftharpoons \text{antigorite} + 3 \text{ brucite} \qquad (1)$$

But it apparently does not apply to the later stages of recrystallization. This reaction has been observed in many prograde metamorphic sequences (Dungan, 1977; Trommsdorff and Evans, 1974; Vance and Dungan, 1977). Field observations in the Central Alps (Evans et al., 1976) have shown that the first appearance of antigorite in ultramafic bodies coincides with the prehnite–pumpellyite/pure pumpellyite facies transition in the regionally metamorphosed host pelites, the reaction (1) being
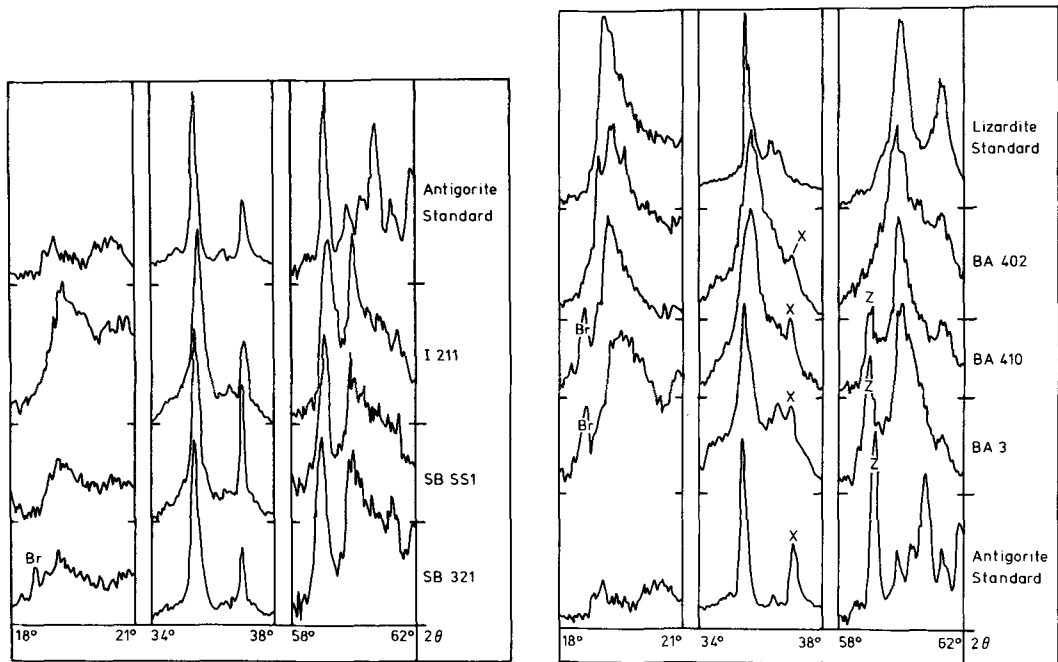
FIGS. 6 and 7. FIG. 6 (*left*). X-ray diffraction patterns of representative samples of non-pseudomorphic serpentinites from Innellan (I) and Scalpsie Bay, Isle of Bute (SB), and the pure antigorite standard. FIG. 7 (*right*). X-ray diffraction patterns of representative samples of 'mixed-type' serpentinites from Balmaha, together with antigorite and lizardite standards. The composite 003 18.00, and 24.30 antigorite reflections are labelled X and Z respectively: see text for discussion. Accessory brucite reflections are labelled Br.

coincident with the beginning of the greenschist facies (300–350 °C, 3–4 kbar).

Wicks and Whittaker (1977) consider that under conditions of rising temperature, the mineral assemblages produced during the metamorphic recrystallization of lizardite serpentinites are of two types: antigorite + brucite, and antigorite alone. Thus, metamorphism of the H.B.S. serpentinites probably occurred close to the boundary between these two regimes, which Wicks and Whittaker (1977) place at a similar temperature to the reaction:

antigorite + 20 brucite $\rightleftharpoons$ 34 forsterite + 51 water
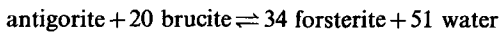
although we have not observed any metamorphic olivine in the non-pseudomorphic serpentinites. If this is the case, then the available data (Evans *et al.*, 1976) indicates metamorphism having taken place at temperatures of 350 ± 50 °C. The transition from an antigorite + brucite assemblage to that of antigorite alone, however, has implications with regard to mass (chemical) balance considerations. Although Wicks and Whittaker (1977) are aware that the conversion of lizardite to antigorite involves the loss of small amounts of $Mg(OH)_2$ (because of the antigorite lattice's deficiency in this

component compared with lizardite), and that in their lower temperature regime this results in the production of brucite, they do not offer an explanation for the fate of the excess $Mg(OH)_2$ in the higher temperature (brucite-absent) regime. In the absence of any further data, we suggest that in the higher temperature (antigorite-only) regime, the excess $Mg(OH)_2$ was removed in solution in the metamorphic fluids; but there is obviously a need to make further studies of the mineralogical and chemical mass balance effects of the lizardite → antigorite transition in metamorphosed serpentinites.

Regardless of these problems, however, it is clear from the textural and mineralogical evidence that the antigorite serpentinites are the (greenschist facies) metamorphosed counterparts of the lizardite serpentinites. The metamorphic status of these rocks is confirmed by stable-isotope data (Ikin and Harmon, 1981, 1983) which show that the antigorite serpentinites exchanged with metamorphic-type fluids, isotopically similar to the Dalradian (Grampian) regional metamorphic waters.

Post-metamorphic processes. The pseudomorphic, mixed-type, and non-pseudomorphic rocks at each of the five sample localities, are all traversed

by cross-fibre chrysotile veins. At Scalpsie Bay, Innellan, and Balmaha such veining is clearly post-metamorphic and it seems likely that the veining at Alyth and Carity Burn, Kirriemuir was contemporaneous with that at the other three localities.

At Cortachy Bridge (Kirriemuir), Scalpsie Bay, and Innellan, considerable, but localized, shearing along discrete zones produced platy talc-chrysotile shear planes. In the case of the rocks at Innellan and Scalpsie Bay, the physical breakdown of the antigorite serpentinites during shearing was a product of retrogressive dynamic metamorphism according to the reaction:

$$\text{antigorite} \rightleftharpoons 15 \text{ chrysotile} + \text{talc}$$
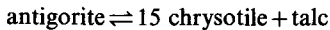
which has not been determined experimentally but, on empirical evidence, has been placed at c. 200 °C (Evans et al., 1976). The relative SiO_2-deficiency of chrysotile with respect to antigorite accounts for the production of talc. However, the same cannot be true for the production of talc and chrysotile from lizardite at Cortachy Bridge, where the introduction of SiO_2-rich fluids is required.

This 'ductile' shearing of the serpentinites contrasts with the more 'brittle' type of deformation which resulted in fracturing and chrysotile vein-formation. Under a single set of P-T conditions, ductile, followed by brittle, deformation could have resulted from tectonism that occurred at progressively higher strain rates; however, stable-isotope evidence suggests an alternative explanation. The chrysotile veins from the antigorite serpentinites at Innellan have H-isotope compositions indicative of their formation from meteoric waters, whereas the sheared lizardite serpentinites from Cortachy Bridge (Kirriemuir) partially exchanged with non-meteoric fluids (Ikin and Harmon, 1983). This indicates that the former were produced at higher crustal levels than the latter. Therefore, with Raleigh and Paterson (1965) having shown that the response of serpentine to deformation changes at c. 2 kbar load pressure (c. 5 km) from brittle to ductile, we consider it more likely that these two forms of deformation reflect tectonism at different crustal levels (and consequently P-T conditions) rather than varying strain rates at relatively uniform P-T conditions.

Conclusions. The variation in mineral assemblages in the Highland Border Suite serpentinites records a complex geological history. The main conclusions from this paper are as follows:

(i) The Highland Border Suite ultramafic rocks are the remnants of a chemically depleted harzburgite mass containing concordant dunite layers and discordant orthopyroxenite dykes and segregations. These rocks are conjectured to be the depleted mantle left after melting of primary lherzolite that produced the basaltic rocks of the Suite.

(ii) Low-temperature serpentinization of the peridotites produced pseudomorphic lizardite serpentinites. The minor-element 'fingerprints' of the primary phases were inherited by the lizardite. Olivine was the first phase to be altered, followed by orthopyroxene and the simultaneous peripheral alteration of Cr-spinel to magnetite. This latter process released Cr and Al cations which were incorporated into the bastite lizardite, thereby enriching it in these two elements above the levels present in the orthopyroxene.

(iii) The lizardite serpentinites were subsequently subjected to greenschist-facies metamorphism; they recrystallized outside the P-T stability field of lizardite, and were converted to non-pseudomorphic antigorite serpentinites.

(iv) Post-metamorphic brittle fracturing resulted in cross-fibre chrysotile vein-formation. This occurred at a relatively high (< 5 km) crustal level where meteoric waters had access.

(v) Post-metamorphic ductile deformation, in the form of talc–chrysotile shear zones, occurred at deeper levels and at temperatures of c. 200 °C with the interaction of (non-meteoric) metamorphic-type fluids.

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