

## Chromite-chlorite intergrowths in peridotite at Chimwadzulu Hill, Malawi

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**SUMMARY.** Chromiferous spinel and chlorite are associated and sometimes intergrown in serpentinized peridotites at Chimwadzulu Hill. The peridotites probably crystallized from a melt under the conditions of the spinel-lherzolite facies (O'Hara, 1967) in which the stable aluminous phase is spinel. The spinel that formed was probably a chromite with some  $\text{Al}_2\text{O}_3$  and  $\text{MgO}$ . At lower temperatures aluminous spinel is unstable in the presence of olivine and orthopyroxene, chlorite being the stable aluminous mineral. It is thought that the chlorite crystallized under the conditions of medium-grade metamorphism that prevailed when the rocks were emplaced, and at the same time the spinel re-crystallized, depleted in  $\text{Al}_2\text{O}_3$  and  $\text{MgO}$  and correspondingly enriched in  $\text{FeO}$ ,  $\text{Fe}_2\text{O}_3$ , and  $\text{Cr}_2\text{O}_3$ , giving rise to the observed chromite-chlorite association.

THE Chimwadzulu Hill ultrabasic body (Bloomfield, 1958) lies at  $15^\circ 02' \text{S } 34^\circ 39\frac{1}{2}' \text{E}$ , about 80 km WNW of Zomba in southern Malawi. It covers about  $0.7 \text{ km}^2$ , and is surrounded by gneisses and schists of the Malawi Basement Complex (Bloomfield and Garson, 1965). The body is composed of three rather variable rocks, the most abundant being hornblende-rich rock, commonly with epidote and more rarely with plagioclase, iron oxides, clinopyroxene, clinozoisite, garnet, corundum, sphene, spinel, and orthopyroxene. Irregularly intermingled with the above are areas of tremolite, amphibolite, and serpentinized peridotite. The tremolite is generally associated with orthopyroxene and magnetite but less commonly with orthoamphibole. The serpentinized peridotite is the most homogeneous of the rock types, consisting of olivine and orthopyroxene both partially replaced by serpentine. An opaque spinel is an ubiquitous accessory; chlorite is a frequent constituent; talc, carbonate, colourless clinoamphibole (probably tremolite), green spinel with opaque inclusions, brown spinel, and hematite with exsolved ilmenite occur locally; and in one specimen Bloomfield (1958) recognized titanclinohumite.

*Chromite-chlorite intergrowths.* Chlorite occurs in the serpentinized peridotites as well-formed crystals showing grey polarization colours. It occurs close to grains of opaque spinel and in some specimens intergrowths of the two minerals are observed. The texture suggests that the two minerals crystallized simultaneously and that the serpentine formed at a later stage, contrary to Bloomfield's (1958) suggestion that the chlorite formed later than the serpentine. The latter mineral fills cracks in the spinel and is clearly late, whereas the manner in which chlorite plates penetrate the spinel

TABLE I. *Electron probe analyses of opaque spinels. Anal. A. E. Tresham*

	A	B	C	D	E1	E2	F	G	I	H	J
SiO <sub>2</sub>	0.9	0.3	0.0	0.1	0.0	0.0	0.0	0.3	0.2	0.3	3.3
TiO <sub>2</sub>	0.3	0.4	0.2	0.3	0.2	0.4	0.0	0.0	0.0	0.0	0.0
Al <sub>2</sub> O <sub>3</sub>	3.5	0.0	6.0	3.5	7.1	3.6	1.2	1.0	1.0	1.8	1.3
Fe <sub>2</sub> O <sub>3</sub> *	19.2	34.3	19.4	20.7	18.1	22.0	42.0	44.2	48.0	42.1	41.2
FeO*	25.3	28.0	22.2	24.0	22.6	23.8	23.4	24.9	25.0	25.0	21.7
MnO	0.7	0.6	0.6	0.8	0.6	0.7	0.5	0.5	0.4	0.5	0.5
MgO	4.8	1.6	6.2	5.1	6.3	4.9	4.6	3.9	3.5	3.8	8.8
CaO	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
NiO	0.4	0.8	0.4	0.4	0.3	0.5	0.6	0.7	0.8	0.7	0.5
CoO	0.2	0.1	0.1	0.1	0.2	0.1	0.1	0.1	0.1	0.1	0.2
ZnO	0.2	0.2	0.3	0.1	0.3	0.2	0.1	0.0	0.1	0.0	0.0
Cr <sub>2</sub> O <sub>3</sub>	45.5	32.9	45.0	45.8	45.7	44.4	27.9	25.1	20.9	25.5	23.7
V <sub>2</sub> O <sub>3</sub>	0.1	0.0	0.1	0.2	0.1	0.2	0.1	0.0	0.0	0.1	0.1
Total	101.3	99.5	100.7	101.1	101.6	100.8	100.5	100.7	99.8	99.9	101.1
Unit cell (on 32 oxygen atoms and 24 cations)											
Si	0.25	0.10	0.00	0.04	0.00	0.00	0.00	0.08	0.05	0.09	0.91
Ti	0.07	0.10	0.05	0.07	0.05	0.08	0.00	0.00	0.01	0.00	0.00
Al	1.17	0.01	1.97	1.16	2.29	1.21	0.42	0.34	0.33	0.61	0.41
Cr	10.11	7.82	9.86	10.18	9.86	9.93	6.39	5.78	4.88	5.90	5.18
V	0.03	0.00	0.03	0.04	0.03	0.04	0.02	0.01	0.00	0.03	0.01
Fe <sup>3+</sup>	4.05	7.77	4.05	4.39	3.72	4.67	9.17	9.72	10.68	9.29	8.58
Fe <sup>2+</sup>	5.94	7.05	5.15	5.64	5.15	5.63	5.69	6.09	6.18	6.12	5.01
Mn	0.17	0.16	0.15	0.19	0.15	0.17	0.11	0.12	0.09	0.12	0.11
Mg	2.03	0.73	2.58	2.13	2.58	2.07	2.00	1.69	1.55	1.65	3.63
Ca	0.00	0.00	0.00	0.00	0.01	0.01	0.00	0.00	0.00	0.00	0.00
Ni	0.09	0.18	0.08	0.10	0.08	0.11	0.15	0.16	0.18	0.16	0.11
Co	0.05	0.03	0.03	0.03	0.03	0.03	0.02	0.02	0.02	0.03	0.04
Zn	0.05	0.05	0.06	0.01	0.06	0.04	0.03	0.00	0.02	0.01	0.01
Olivine	0.75	0.31	0.00	0.12	0.00	0.00	0.00	0.23	0.14	0.26	2.74
Ulvösp.	0.22	0.29	0.14	0.22	0.14	0.23	0.00	0.00	0.03	0.00	0.00
Spinel	23.03	23.40	23.86	23.66	23.85	23.77	24.00	23.77	23.83	23.73	21.26
Mg/(Mg+Fe <sup>2+</sup> )	0.25	0.09	0.33	0.27	0.33	0.27	0.26	0.22	0.20	0.21	0.42
Cr/(Cr+Al)	0.90	1.00	0.83	0.90	0.81	0.89	0.94	0.94	0.94	0.91	0.93
Fe <sup>3+</sup> /(Fe <sup>3+</sup> +Cr+Al)	0.26	0.50	0.25	0.28	0.23	0.30	0.57	0.61	0.67	0.59	0.61

\* Calculated assuming the ideal formula  $R^{2+}R_2^{3+}O_4$ .

A to E2 in a peridotite (Geol. Surv. Malawi no. HH53, I.G.S. no. F7793), containing about 40 % olivine, 40 % enstatite, 2 % opaque spinel (often with adjacent flakes of chlorite), and traces of sulphide. The rest of the rock is made up of serpentine, talc, and carbonate. Identifications of chlorite and talc were confirmed by X-ray powder photographs.

F to J in a serpentinized peridotite (Geol. Surv. Malawi no. HH26, I.G.S. no. F7792), consisting of angular fragments of fresh olivine and enstatite resting in a mesh structure of pale-yellow chrysotile. Small grains of opaque spinel make up less than 2 % of the rock and are sometimes associated with flakes of chlorite. There is a small amount of carbonate. The identifications of chlorite and chrysotile were confirmed by X-ray powder photographs.

has some resemblance to plagioclase-augite relationships in the ophitic texture of dolerites and suggests simultaneous crystallization.

These chromite-chlorite associations are thought to have formed by the expulsion of Mg and Al from a high-temperature spinel phase as the rock cooled from the

TABLE II. *Analyses of chlorites in serpentinized peridotites*

	1	2	3		1'	2'	3'		
SiO <sub>2</sub>	31.51	32.17	31.87	Si	6.07	8.00	6.16	6.08	8.00
TiO <sub>2</sub>	0.07	0.03	0.17	Al	1.93		1.84	1.92	
Al <sub>2</sub> O <sub>3</sub>	15.43	14.24	14.51	Al	1.57	12.08	1.38	1.34	11.74
FeO*	2.34	2.28	5.25	Ti	0.01		0.01	0.02	
MnO	0.03	0.03	tr	Fe	0.38	0.37	0.81		
MgO	34.32	34.94	32.76	Mn	0.01	0.01	—	0.03	
CaO	—	—	—	Mg	9.85	9.98	—	9.38	
Na <sub>2</sub> O	0.01	—	0.04	Cr	0.21	0.31	—	0.16	
K <sub>2</sub> O	0.01	—	0.02	Ni	0.05	0.03	—	0.03	
H <sub>2</sub> O	—	—	13.95	OH	16.00	16.00	16.00	16.62	16.62
Cr <sub>2</sub> O <sub>3</sub>	1.35	2.03	1.10						
NiO	0.35	0.18	0.28						
Total	85.42	85.90	100.13						

1. Chlorite flake adjacent to spinel grain C (see Table I) in specimen HH53 (F7793). Anal. A. E. Tresham.

2. Chlorite flake adjacent to a spinel grain that was not analysed in specimen HH26 (F7792) (see Table I). Anal. A. E. Tresham.

3. Chromiferous clinocllore from serpentinite from Kaukapakapa, New Zealand (Hutton and Seelye, 1947).

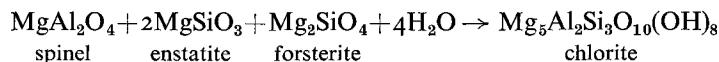
1', 2', 3'. Atomic ratios to O<sub>28</sub> (OH)<sub>16</sub>.

\* Total iron is quoted as FeO.

conditions of the spinel-lherzolite facies to those of the chlorite-amphibole-peridotite facies.

The spinel-lherzolite facies (O'Hara, 1967) is characterized by the assemblage forsterite+diopside+enstatite+spinel. It is the stable assemblage in peridotites crystallizing between 8 and 18 kb being stable down to about 950 °C. At temperatures between about 720 and 820 °C and over a wider range of pressures, aluminous spinel is unstable in the presence of olivine, enstatite, and water vapour, the stable aluminous phase being magnesian chlorite (Fawcett, 1964). This is the chlorite-amphibole-peridotite facies (O'Hara, 1967).

It is suggested, therefore, that the Chimwadzulu Hill peridotites crystallized from a magma under the conditions of the spinel-lherzolite facies, and subsequently recrystallized in the chlorite-amphibole-peridotite facies. The aluminous spinel that crystallized under the former conditions was unstable at the lower temperatures of the latter. Alumina and magnesia were released from the spinel lattice and combined with olivine, enstatite, and water vapour to form chlorite:



This was accompanied by the recrystallization of the spinel phase, richer in iron and chromium. The growth of these chromite-chlorite intergrowths containing some large well-formed chlorite crystals, and the development in some peridotite specimens of large crystals of tremolite, show that the rocks were maintained for a substantial period of time at the conditions of the chlorite-amphibole-peridotite facies. This may

be correlated with the almandine-amphibolite facies assemblages developed in the surrounding gneisses (Bloomfield and Garson, 1965), and it is suggested that the ultrabasic rocks were intruded syntectonically into these gneisses, crystallizing initially as a high-temperature assemblage at the temperature of solidification and subsequently recrystallizing in the lower temperature assemblage after cooling to the prevailing conditions of regional metamorphism. The peridotites continued to cool, as the temperature of the surrounding gneisses decreased, permitting the widespread development of serpentine at temperatures below about 500 °C and the more local formation of talc and magnesite. Olivine and orthopyroxene were extensively, though variably, replaced in this final stage, but chromite and chlorite survived.

*Electron-microprobe analyses.* Analyses of spinel and associated flakes of chlorite in two samples of serpentinized peridotite are given in tables I and II. The spinel analyses were recalculated into unit cell formulae, assigning Si to olivine, Ti and ulvöspinel ( $\text{Fe}_2\text{TiO}_4$ ), and the remaining elements to spinel. The amounts of FeO and  $\text{Fe}_2\text{O}_3$  were calculated assuming the divalent and trivalent elements in the spinel to be present in the ratio 1:2. The unit cells of the chlorites were calculated assuming  $\text{OH} = 16.00$  and 28 remaining oxygen equivalents. Compositional plots of the spinels (fig. 1) show that the spinel in HH53 (F7793) is a ferrian chromite, while in HH26 (F7792) it is chromian magnetite.

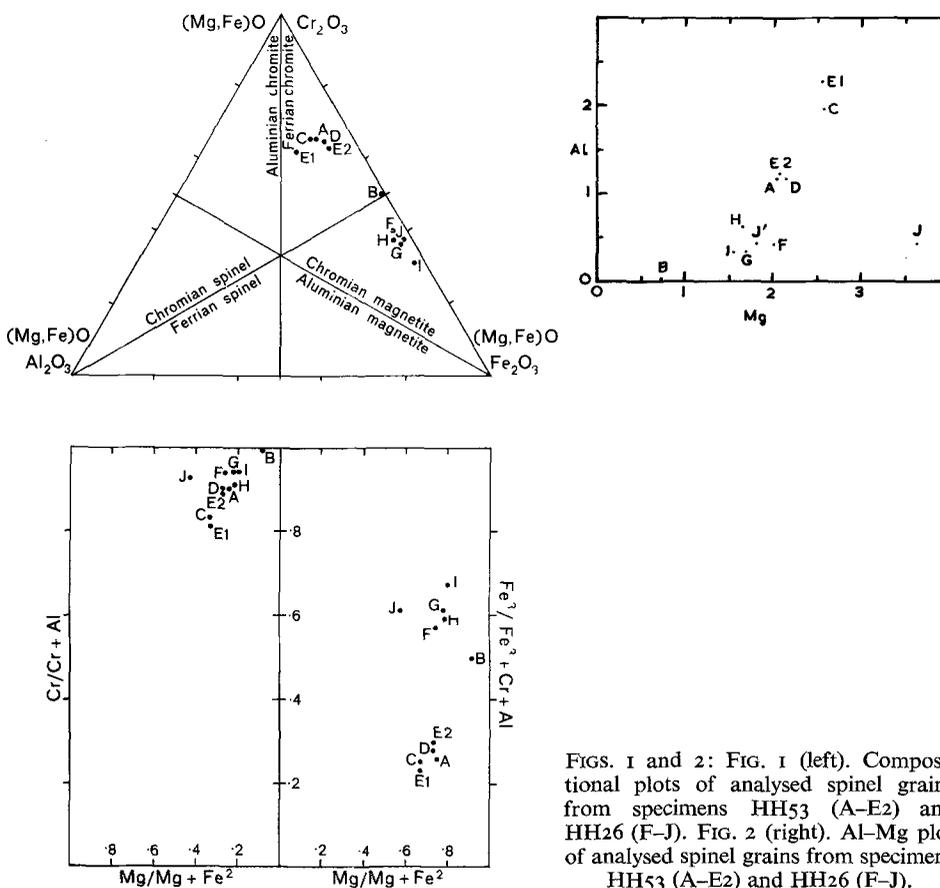
In specimen HH53 (F7793), grains A, C, D, E1, and E2 (Table I) are adjacent to flakes of chlorite and are considered to have lost  $\text{Al}_2\text{O}_3$  and MgO. There is some variation in the  $\text{Al}_2\text{O}_3$  and MgO contents, and they appear to be related (fig. 2). A detailed probe traverse of grain E1 confirmed the variations in  $\text{Al}_2\text{O}_3$  content showing 8.5 %  $\text{Al}_2\text{O}_3$  and 37 % FeO (total iron) at the core, zoned outwards to a rim with 5 %  $\text{Al}_2\text{O}_3$  and 42 % FeO (total iron). Grain E2 did not have regular zoning but again showed variations in the  $\text{Al}_2\text{O}_3$  content. Grain B is not, in the plane of the section, associated with chlorite and its composition differs from the others in its higher content of total iron (FeO and  $\text{Fe}_2\text{O}_3$ ) and lower  $\text{Cr}_2\text{O}_3$ , MgO, and  $\text{Al}_2\text{O}_3$ . It may perhaps be a high-temperature non-aluminous phase that retained its original composition on cooling.

In specimen HH26 (F7792) only grain F is associated with chlorite in the plane of the section, but four of the five analysed grains plot near the same line in fig. 2 ( $\text{Al} = 2(\text{Mg} - 1.5)$ ) as the grains from the other specimen. Grain J is a very small grain enclosed by olivine, and the high content of olivine in the unit cell as calculated from the analysis is probably due to contamination. If the olivine component is subtracted from the analysis, the revised point J' in fig. 2 falls on the same line as the other grains.

The analysed chlorites from the same two samples are clinoclhores containing significant  $\text{Cr}_2\text{O}_3$  (Table II). They compare quite closely with a chromian clinocllore from a serpentinite at Kaukapakapa, New Zealand. The Cr reflects the origin of the chlorites, formed in association with a chrome-rich spinel, but it is curious that the chlorite in sample HH 26 (F7792) has more  $\text{Cr}_2\text{O}_3$  than that in HH53 (F7793), while the reverse is true of the spinels.

*Discussion.* Chromiferous spinels, somewhat similar in composition to those at Chimwadzulu Hill, have been described from the Stillwater Complex, Montana, and

the ultramafic deposits of the Pennsylvania-Maryland State Line district. At each locality a low-alumina spinel was formed in association with an aluminous chromite during the serpentinization of the surrounding rocks.



FIGS. 1 and 2: FIG. 1 (left). Compositional plots of analysed spinel grains from specimens HH53 (A-E2) and HH26 (F-J). FIG. 2 (right). Al-Mg plot of analysed spinel grains from specimens HH53 (A-E2) and HH26 (F-J).

In the Stillwater Complex, Beeson and Jackson (1969) described a ferrit-chromit rim (with 3 to 7 %  $\text{Al}_2\text{O}_3$  and 2 %  $\text{MgO}$ ) surrounding grains of chromite (with 19 %  $\text{Al}_2\text{O}_3$  and 9 to 10 %  $\text{MgO}$ ). The rims were formed from the parent chromite by volume-for-volume replacement, during which  $\text{Al}_2\text{O}_3$  and  $\text{MgO}$  were replaced by  $\text{Fe}_2\text{O}_3$  and  $\text{FeO}$  while the  $\text{Cr}_2\text{O}_3$  content remained nearly constant. This alteration appears to be genetically related to the formation of chlorite, and is likely to have taken place at the same time as dunites above and below were serpentinized.

In the ultramafics of the Pennsylvania-Maryland State Line district, grains of chromite typically containing 9 %  $\text{Al}_2\text{O}_3$  and 9 %  $\text{MgO}$  are commonly rimmed by ferrit-chromit containing 0.3 %  $\text{Al}_2\text{O}_3$  and 3.3 %  $\text{MgO}$  (Ulmer, 1974). In contrast to

the Stillwater occurrence, there are no associated aluminous silicates and it was suggested that the ferrite-chromite is a secondary overgrowth resulting from the aqueous mobilization of chromium during serpentinization.

In the Chimwadzulu Hill specimens there is no sign of one phase rimming another. Chlorite is present as in the Stillwater specimens, and its formation is clearly related to that of the spinel phase. The two minerals are sometimes intergrown in a manner not described at Stillwater and suggesting simultaneous crystallization. The conversion of aluminous chromite to a low-alumina chromite in the Stillwater Complex is considered to have taken place at the temperature of serpentinization. While it may not be impossible that the reaction took place under the same conditions at Chimwadzulu Hill, there are some features that suggest a higher temperature:

1. The reaction has proceeded almost to completion (not entirely as the zoning in grain E1 shows), whereas at Stillwater only the outer rims of the grains have been converted.
2. At Stillwater the original crystal outlines are preserved, whereas at Chimwadzulu Hill the intergrowths point to a more thorough recrystallization.
3. The presence and abundance of chlorite are not related to the degree of serpentinization of the rock.
4. The existence of mineral assemblages of the almandine-amphibolite facies in the surrounding gneisses and in the associated amphibolites of the ultrabasic mass, and the presence of amphibole in one specimen of the serpentinized peridotite show that the rocks were maintained at temperatures of about 650 to 800 °C for long enough for mineral assemblages unstable at these conditions to be converted to stable assemblages. O'Hara's (1967) data indicate that aluminous spinel is not stable and that it would be replaced by chlorite.

It is therefore considered that the chlorite and associated chromiferous spinel crystallized together under the conditions of the chlorite-amphibole-peridotite facies. It is possible, however, that there was minor adjustment of the compositions of the minerals at lower temperatures. For example, the lower  $\text{Al}_2\text{O}_3$  content of the spinel in the more highly serpentinized rock (HH26) might be the result of further reaction at the temperature of serpentinization.

There is little evidence of the nature of the primary aluminous spinel. A green spinel with opaque inclusions occurs in one peridotite specimen, and this is a possible relict of the primary spinel. However, the relative abundances of chlorite and its associated spinel suggest that the original mineral was richer in iron and chromium, and it was probably a chromite with substantial quantities of alumina and magnesia.

*Conclusions.* The Chimwadzulu Hill ultrabasic rocks were emplaced syntectonically during a period of almandine-amphibolite facies metamorphism. The peridotites crystallized from a magma under the conditions of the spinel-lherzolite facies, and one of the minerals to crystallize was an aluminous chromite. When the rocks had cooled to the temperature of the surrounding regional metamorphism, the peridotites recrystallized with assemblages of the chlorite-amphibole-peridotite facies:  $\text{Al}_2\text{O}_3$  and MgO were expelled from the spinel and combined with forsterite, enstatite, and water

vapour to form chlorite, while the spinel itself recrystallized as a ferrian chromite or chromian magnetite.

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