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CHEMICAL VARIATION OF CHROMITE IN THE ULTRAMAFIC CUMULATES OF THE GREAT SERPENTINITE BELT, UPPER BINGARA TO DOONBA, NEW SOUTH WALES, AUSTRALIA

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ABSTRACT

Compositional variation for disseminated chromite in olivine-dominated cumulate rocks from the ophiolite sequence of the Great Serpentinite Belt, in New South Wales, Australia, is best displayed by divalent cations. $X(\mathrm{Fe}^{2+})$ of chromite included in cumulus olivine is higher than that of chromite found entirely within postcumulus plagioclase and clinopyroxene. These variations result from subsolidus re-equilibration between chromite and olivine at the grain scale and give an inverse correlation between $X(\mathrm{Fe}^{2+})$ and grain size of the chromite. Chromite included entirely within clinopyroxene is similar in composition to that located entirely in plagioclase; both types are largely unaffected by subsolidus reactions. A range of $X(\mathrm{Fe}^{2+})$ for intergrain chromite located entirely within plagioclase, which is nearly twice as large as that shown by chromite associated with olivine, is inherited from the magmatic stage. Chemical differences between concentrated and disseminated types of chromite reflect initial differences in conditions of crystallization for the two types.

Keywords: chromite, chromitite, cumulate rocks, subsolidus reactions, ophiolite sequence, Great Serpentinite Belt, Australia.

SOMMAIRE

La variation en composition de la chromite disséminée dans des cumulats d'olivine de la séquence ophiolitique dite de Great Serpentinite Belt, au New South Wales, en Australie, est surtout exprimée par les cations bivalents. La valeur de $X(\mathrm{Fe}^{2+})$ de la chromite incluse dans l'olivine cumulative est supérieure à celle de la chromite incluse dans le plagioclase et le clinopyroxène post-cumulus. Ces variations résultent d'un ré-équilibrage subsolidus entre la chromite et l'olivine à l'échelle des grains, et mène à une corrélation inverse entre $X(\mathrm{Fe}^{2+})$ et la taille des grains de chromite. La composition de la chromite entièrement piégée par le clinopyroxène est semblable à celle qui est entièrement incluse dans le plagioclase; dans les deux cas, les réactions subsolidus n'ont exercé aucune influence importante. L'étendue des valeurs de $X(\mathrm{Fe}^{2+})$ parmi les grains de chromite situés entièrement à l'intérieur du plagioclase, presque le double de la variabilité dans les grains de chromite associés à l'olivine, serait héritée du stade magmatique. Les différences chimiques entre chromite concentrée dans des chromitites et chromite disséminée résulteraient de différences initiales en conditions de cristallisation dans les deux milieux.

(Traduit par la Rédaction)

Mots-clés: chromite, chromitite, roches cumulatives, réactions subsolidus, séquence ophiolitique, Great Serpentinite Belt, Australie.

INTRODUCTION

Chromite is commonly present as an accessory mineral in ultramafic rocks. Its composition is quite sensitive to the physicochemical conditions of magmatic systems; thus its chemistry can be used to indicate the nature and evolution of magmatic processes during the course of its crystallization (Irvine 1965, 1967). Compositional variation during initial crystallization of chromite can result from variables at the magmatic stage, above the solidus. The composition of magmatic chromite may change by reaction between early-formed chromite and the evolving magma, for example between chromite incorporated in cumulus layers and interstitial melt. In addition, its composition may be modified at the subsolidus stage, as a result of cation exchange between chromite and associated silicate phases in the presence of a fluid medium. The distinction between chromite formed during the magmatic interval and chromite affected by subsolidus reactions is important in the identification of processes that are operating at each stage, and can commonly be made on textural and chemical grounds.

The composition of the chromite in a particular body of rock may vary significantly, in response to lithological changes or the nature of associated silicate minerals. The dependence of the composition of chromite on the nature of the associated silicate minerals has been observed by many investigators, including Cameron (1975), Hamlyn & Keays (1979), Henderson & Wood (1981), Wilson (1982), Hatton & von Gruenewaldt (1985), and Roeder & Campbell (1985). Usually, chromite is classified on the basis of compositional variation into one of two groups: (i) that included in cumulus olivine, and (ii) that encountered in postcumulus plagioclase and pyroxene. However, chemical differences between the two groups of chromite can result from reactions taking place either above or below the solidus.

To determine the cause of these chemical variations, it is important to characterize the types of chromite on the basis of associated silicate minerals, especially in the common case where critical textural evidence for solid–solid, or solid–liquid reaction, is lacking. The ultramafic cumulates of the Great Serpentinite Belt, New South Wales, provide a variety of occurrences of chromite that have the imprints of both magmatic and subsolidus processes. By comparing differing textural types of chromite and their associated minerals, this study aims firstly, to investigate the effect on chromite composition of subsolidus re-equilibration between chromite and adjacent silicate minerals, and secondly, to assess the cause for the compositional variation in chromite prior to the subsolidus stage.

GEOLOGICAL SETTING AND OCCURRENCE OF CHROMITE

An Alpine-type association of serpentinized harzburgite, ultramafic cumulate, gabbro, dolerite, basalt and minor plagiogranite is distributed along the Great Serpentinite Belt, in the southern portion of the New England Fold Belt in eastern Australia. The Great Serpentinite Belt is confined to the Peel Fault Zone (Fig. 1), which forms a vertical or steep westerly dipping crustal suture, representing a long-lived Paleozoic convergent plate margin (Offler & Williams 1987). To the west of the Peel Fault Zone lies a fore-arc basin sequence (Tamworth Belt), comprising a Paleozoic regressive marine to terrestrial, volcanic–sedimentary succession. To the east of the fault, a subduction sequence (Central Block) consists mainly of Paleozoic deep marine sediments overlain by Paleozoic to Tertiary volcanic and fluvial sequences (Brown 1987). A variation in the grade of metamorphism is recorded in the sequences on each side of the fault. Diagenetic to lower anchizonal (subgreenschist) grades are encountered in the Tamworth Belt, and diagenetic to epizonal (subgreenschist to greenschist) grades are recognized in the Central Block (Offler & Hand 1988).

The rock association in the Great Serpentinite Belt most likely represents a dismembered ophiolite sequence (Crook & Felton 1975, Pooley 1979, Rogers 1986). In the region from Upper Bingara to Doonba in northern New South Wales (Fig. 1), serpentinized harzburgite and gabbro are the major exposed lithologies in the association. Dolerite sills and basalt represent less abundant rock-types, confined to an area east of the gabbro complex, or to the contact between gabbro and harzburgite. Minor plagiogranite dykes are distributed in both harzburgite and gabbro. Exposure is variable over the area, and ranges from as high as 70% over the harzburgite to about 15% over gabbro, dolerite and basalt.

Deformation has intensely disturbed the sequence, resulting in faulted contacts between most members of the association and a reduction in the thickness of the whole sequence to about 2 km, which is considerably thinner than typical oceanic crust (7–8 km) and other recognized ophiolite sequences. The presence of additional faults within individual units is suspected, but this is difficult to establish because of the subdued relief on many outcrops or lack of exposure.

Chromite is encountered in ultramafic cumulates and the serpentinized harzburgite. Cumulates, mostly comprising lens-like units, lie along the base of a NNW-trending gabbro complex (Fig. 1). Outcrops of the cumulates range up to about 200 m in length and 50 m in width and demonstrate long axes nearly parallel to the contact between the gabbro complex and harzburgite. The gabbro has a mineralogy dominated by plagioclase (48–55 modal %) and clinopyroxene (45–52%), but lacks chromite.

Mineral modes for individual bodies of cumulates are quite uniform, and comprise 60–75% olivine, 23–25% clinopyroxene + plagioclase, and 0.5–4% chromite. Euhedral to subhedral cumulus olivine, with a grain size of 1.2–8 mm, forms the mineral framework (Fig. 2a). These olivine-dominant rocks are termed olivine–chromite cumulates. Adcumulus overgrowth of olivine, usually toward the lower portion of the ultramafic sequence, results in a rock approaching a dunitic composition, with at least 85 modal % olivine. Texturally, a simple order of crystallization for the olivine–chromite cumulate is evident: olivine + chromite (cumulus phases) were followed by clinopyroxene + calcic plagioclase (postcumulus).

Calcic plagioclase and clinopyroxene fill interstitial spaces between olivine (Fig. 2a). Clinopyroxene commonly demonstrates a poikilitic habit and cotectic crystallization with plagioclase (Fig. 2b). Among the different bodies of cumulates, the modal abundance of plagioclase and clinopyroxene may range from 10 to 34% and from 2 to 15%, respectively, representing a variation in rock type from plagioclase wehrlite to troctolite.

Cumulus chromite is dispersed throughout the entire volume of ultramafic cumulates. The grain size of the disseminated chromite is considerably smaller than that of the associated olivine, and lies in the range 35 to 600 μm in diameter, with most grains occupying the size

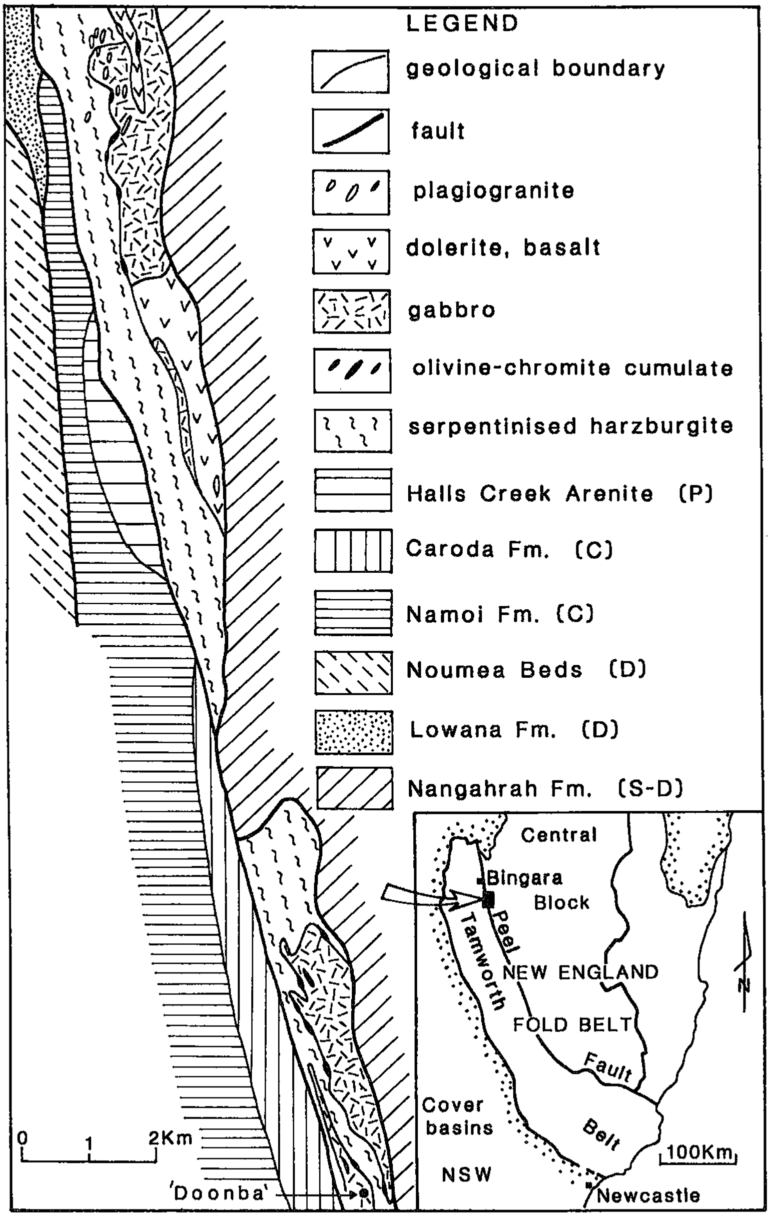

FIG. 1. Geological map and location of the Great Serpentinite Belt from Upper Bingara to Doonba, New South Wales. The dimensions of olivine–chromite cumulates and plagiogranites are enlarged for greater clarity.

interval between 50 and 400 μm. Chromite interstitial to olivine ("intergrain chromite") is more abundant than chromite included in olivine ("included chromite"). Whereas the range in grain size of the intergrain chromite varies considerably (40–600 μm), that of included chromite is relatively small (35–300 μm).

Figure 2a displays many of the characteristics of the different textural types of chromite. Some anhedral intergrain crystals occupy and acquire the shape of the entire gap between olivine grains, or between olivine and postcumulus minerals, suggesting crystallization from interstitial melt, either from a new nucleus or by

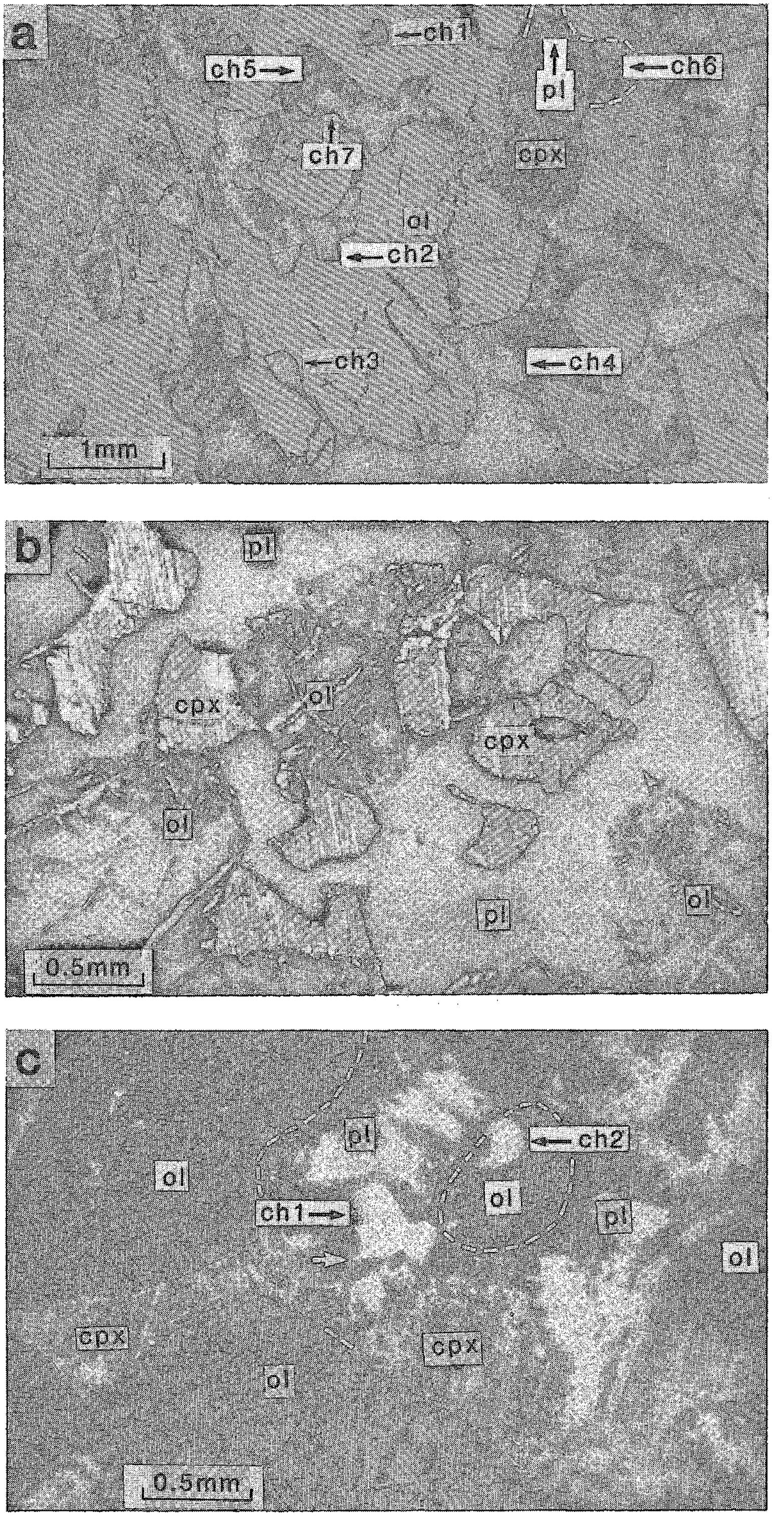

FIG. 2. a) Postcumulus clinopyroxene (cpx) and plagioclase (pl) interstitial to olivine (ol). Chromite (ch) is located both in olivine and postcumulus minerals. Olivine is completely altered to serpentine minerals, and plagioclase is altered to an aggregate of grossular, hydrogrossular and chlorite. Secondary magnetite is distributed along fractures or grain boundaries. Key to numbered chromite grains: (i) included chromite: ch1: entirely within olivine; ch2: in olivine, partly in contact with plagioclase; (ii) intergrain chromite: ch3: between two grains of olivine; ch4: in clinopyroxene, partly in contact with olivine; ch5: entirely within plagioclase; ch6: entirely within plagioclase, (olivine–plagioclase boundary is enhanced); ch7: in plagioclase, partly in contact with olivine. Plane-polarized light. b) Clinopyroxene poikilitically including olivine and plagioclase. Isotropic areas mainly consist of grossular plus hydrogrossular replacing plagioclase. Crossed polars. c) A "tail" (arrow) from an intergrain chromite (ch1) extends between clinopyroxene and plagioclase. Dusty, semi-opaque areas on altered plagioclase are aggregates of andradite, which are surrounded by chlorite (light-colored region). Ch2: included in olivine, partly in contact with plagioclase. Plagioclase–olivine boundaries are enhanced. Plane-polarized light.

overgrowth on settled cumulus chromite during the postcumulus stage. Furthermore, in some cases, the intergrain chromite shows a "tail" from the central part of the grain to fill the interstices between olivine or postcumulus minerals (Fig. 2a). This tail is obviously the latest overgrowth on the cumulus grain. These observations suggest that disseminated chromite had an extensive history of crystallization, which spanned cumulus to postcumulus stages.

The intergrain chromite is either wholly occluded by postcumulus phases and isolated from olivine (Fig. 2a) or incompletely surrounded by postcumulus minerals and partially in contact with olivine. On the other hand, included grains of chromite are wholly confined to single grains of olivine or incompletely enclosed by olivine and in partial contact with postcumulus phases. Replacement or reaction textures are not evident at the contact between chromite and any of the silicate mineral phases.

In addition to the accessory chromite disseminated in olivine-dominant rocks, chromite concentrates within some of the cumulate lenses to form thin bands, from less than 0.5 cm to 7 cm in thickness, containing 60–65 modal % chromite. These chromitite bands ("concentrated chromite"), usually a few cm apart, best display the layered nature of the host cumulate. Chromite, of euhedral to subhedral shape, with a grain size ranging from 0.2 to 1.2 mm, is the only cumulus mineral. A prominent feature of the lenses is that about half of the chromite grains have one or more silicate inclusions (completely chloritized or serpentinized), similar to the case in chromitites of the Bird River Sill (Ohnenstetter *et al.* 1986).

Alteration of the primary mineralogy of the cumulate rocks is significant and has resulted in almost complete serpentinization of olivine and the destruction of feldspar to produce an assemblage of either grossular + hydrogrossular + chlorite, or andradite + chlorite. Clinopyroxene exhibits the effects of slight alteration to chlorite and tremolite, or rare serpentine. Some grains of disseminated chromite are rimmed by fine-grained aggregates of magnetite or contain microfractures filled by magnetite. Concentrated chromite in chromitite bands is variably replaced by "ferritchromit".

COMPOSITIONS OF CHROMITE

Analytical methods

Outcrop samples of both olivine–chromite cumulate and chromitite bands were collected from five separate cumulate lenses for detailed study by optical microscopy and chemical analysis. Spot analyses were made on 326 grains of disseminated chromite and 114 grains of concentrated chromite (data from only two of the lenses are shown in the following figures for clarity). Analyses were performed with a JEOL JSM 840 scanning electron microscope and a Tracor Northern EDS system, using mineral and synthetic oxide standards and ZAF correction procedures: we used a beam current of 2 μA, an accelerating voltage of 15 kV, 60 seconds counting time and a beam diameter of about 2 μm.

Elements determined were Si, Ti, Al, Cr, V, Mn, Ca, Mg, Zn and total Fe. Raw microprobe totals range from 98.4 to 101% for all analyses. Si, V, Mn, Ca, and Zn are present only in trace amounts (less than 0.5 wt% for each of their oxides); for a majority of the chromite grains, these elements are below detection levels of about 0.1 to 0.2 wt%. Two to four spot analyses were made on each grain; additional spots were analyzed for a small number of grains from each sample to check for compositional zoning. Each datum in the accompanying figures represents the average of all spot analyses determined for an individual grain. A complete listing of the analytical data is available from the authors on request.

FeO and Fe_2O_3 are calculated from total Fe by assuming a slight departure from the ideal structure (R^{2+}:R^{3+} = 1:2) due to the presence of a small amount of the ulvspinel component (Deer *et al.* 1962). Because some chromite grains contain small amounts of SiO_2 (up to 0.48 wt%), allowance is also made in the calculation for the entry of minor Si into the spinel structure. Substitution for Al in those grains containing SiO_2 generates a further departure from the ideal formula in the FeO and Fe_2O_3 calculation. In calculating the cation fraction $X(Fe^{2+})$, Fe^{2+} is derived by subtracting that part of the ferrous iron in the $Fe^{2+}_2(Ti,Si)O_4$ component from total ferrous iron.

Results

The five main chemical components (Cr_2O_3, Al_2O_3, MgO, FeO and Fe_2O_3) are present in the following ranges: 38.8 – 43.4, 21.8 – 27.2, 12.6 – 14.8, 13.4 – 16.5 and 2.9 – 4.9 wt%, respectively, for concentrated chromite, and 38.3 – 46.7, 13.5 – 27, 6.6 – 12.8, 16.9 – 25.3 and 4 – 11.6 wt%, respectively, for disseminated chromite. Selected compositions of chromite are listed in Table 1. Compositional zoning within individual grains was not identified. Moreover, a compositional variation of chromite with stratigraphic height in the cumulate sequence is not recorded.

The intergrain chromite from the olivine–chromite cumulates (Figs. 3, 4) spans a greater range of composition, expressed by $X(Fe^{2+})$, relative to the included chromite (Fig. 5). Intergrain chromite, entirely in either plagioclase or clinopyroxene, is distinctly Fe^{2+}-poor compared with included chromite. In contrast, no significant differences in ratios of trivalent cations between the two groups were noted. For the intergrain chromite, that in plagioclase (Fig. 3) has an almost identical composition to that in clinopyroxene (Fig. 4).

Compared to the disseminated chromite, the concentrated chromite in chromitite bands is richer in Mg^{2+} and Al^{3+} (Fig. 6). The compositional variation observed among the samples of concentrated chromite, however,

TABLE 1. SELECTED RESULTS OF ELECTRON-MICROPROBE ANALYSES OF CHROMITE, ULTRAMAFIC CUMULATES OF THE GREAT SERPENTINE BELT

Related silicate	Ol	Ol	Cpx	Cpx	Pl	Pl	Pl,Ol	Pl,Ol	Cpx,Ol	Cpx,Ol	concentrated chromite		
Diameter (μm)	115	120	260	50	115	290	110	200	200	55	—	—	—
Weight percentage													
SiO_2	0.11	u.d.	u.d.	u.d.	u.d.	0.27	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.
TiO_2	0.82	0.97	0.94	1.30	1.15	1.38	1.45	1.23	1.15	0.69	0.42	0.35	0.42
Al_2O_3	17.92	14.87	19.67	18.53	19.64	17.49	16.29	17.61	18.58	18.95	25.38	24.86	27.18
V_2O_3	0.30	0.42	u.d.	0.21	u.d.	0.42	0.26	0.31	u.d.	0.47	0.37	u.d.	u.d.
Cr_2O_3	44.10	43.94	42.79	41.74	42.51	42.70	45.10	43.80	42.63	41.46	40.19	41.46	38.79
ΣFeO	28.86	31.94	25.78	27.36	24.35	27.37	29.12	28.25	27.41	28.70	18.23	19.50	18.10
MgO	8.40	7.49	10.48	9.50	11.56	9.64	8.16	8.80	8.90	8.28	14.31	13.45	14.80
CaO	u.d.	u.d.	u.d.	0.12	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.
MnO	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.
ZnO	0.12	u.d.	u.d.	u.d.	u.d.	u.d.	0.28	u.d.	u.d.	u.d.	u.d.	u.d.	u.d.
Total	100.63	99.63	99.66	98.76	99.21	99.27	100.66	100.00	98.67	98.55	98.90	99.62	99.29
Cations per 32 oxygens													
Si	0.028					0.069							
Ti	0.158	0.192	0.179	0.252	0.218	0.267	0.282	0.238	0.224	0.135	0.077	0.064	0.076
Al	5.410	4.612	5.863	5.627	5.835	5.304	4.961	5.339	5.672	5.804	7.261	7.125	7.672
V	0.062	0.089		0.043		0.087	0.054	0.064		0.098	0.072		
Cr	8.931	9.141	8.555	8.502	8.471	8.686	9.214	8.908	8.730	8.518	7.713	7.971	7.344
Fe^{3+}	1.226	1.774	1.224	1.324	1.258	1.250	1.207	1.213	1.150	1.310	0.801	0.776	0.832
Fe^{2+}	4.956	5.254	4.228	4.571	3.875	4.639	5.085	4.864	4.788	4.927	2.899	3.189	2.792
Mg	3.207	2.938	3.951	3.648	4.343	3.697	3.143	3.374	3.436	3.207	5.178	4.875	5.283
Ca				0.033									
Mn													
Zn	0.023						0.053						
Total	24.000	24.000	24.000	24.000	24.000	24.000	24.000	24.000	24.000	24.000	24.000	24.000	24.000

u.d.=undetected. —=undetermined. Pl,Ol=in Pl, in contact with Ol. Cpx,Ol=in Cpx, in contact with Ol.

cannot be correlated with the stratigraphic height of individual grains within any one chromitite layer.

The nature of the enclosing or adjacent silicate minerals exerts an influence on the composition of individual grains of chromite. Some crystals of intergrain chromite are not wholly included in postcumulus phases, but are partly in contact with cumulus olivine. The data in Figures 3 and 4 demonstrate systematic, though small chemical differences between those grains that are entirely included in clinopyroxene or plagioclase and those that are incompletely enclosed in the postcumulus phases, but are partly in contact with olivine. In the intergrain chromite in partial contact with olivine, $X(Fe^{2+})$ is similar to that of the included chromite, but is substantially higher than that in chromite entirely in either clinopyroxene (Fig. 4) or plagioclase (Fig. 3).

Significant differences in chemistry do not exist among grains of included chromite, irrespective of whether they are enclosed entirely by olivine, or incompletely included by olivine but in partial contact with plagioclase or pyroxene (Fig. 5). Those grains of included chromite not wholly in olivine and in contact with postcumulus phases could have originated by two distinct processes: (i) they may represent inclusions in olivine formed at the cumulus stage, or (ii) they may have formed originally as intergrain chromite later enclosed by olivine because of overgrowth of the host at the postcumulus stage.

For disseminated chromite, $X(Fe^{3+})$ lies generally in the range from 0.05 to 0.12. This partly overlaps the range in values of 0.04 to 0.08 (Fig. 6) for concentrated chromite.

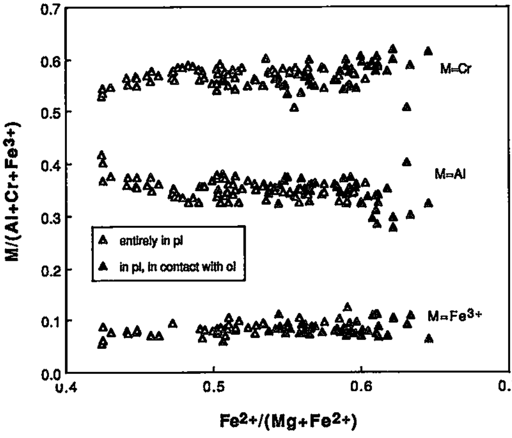

FIG. 3. Composition of intergrain chromite in plagioclase.

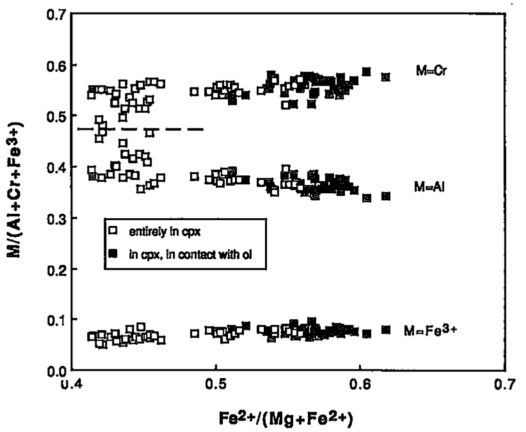

FIG. 4. Composition of intergrain chromite in clinopyroxene.

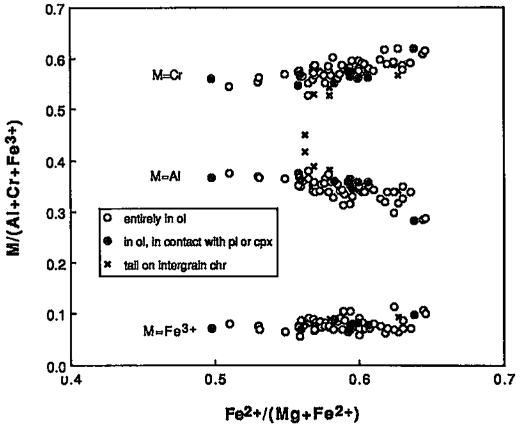

FIG. 5. Composition of included chromite and the overgrowth portion of intergrain chromite.

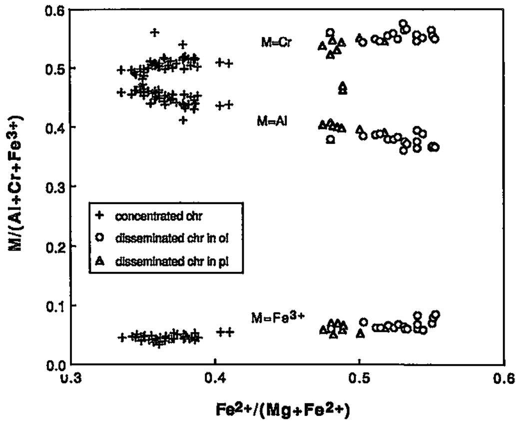

FIG. 6. Composition of concentrated chromite from a chromitite layer and disseminated chromite from adjacent troctolite.

DISCUSSION

Subsolidus equilibria

The compositional variation of chromite in a cumulate can be attributed to any of the following three mechanisms:

1. At the magmatic stage, changes in composition and physicochemical properties of the melt, or reaction between early-formed chromite crystals and evolved silicate liquids, could result in crystallization of chromite with variable composition.
2. At the postcumulus stage, chromite grains in the pile of cumulates may react with the interstitial liquid, with or without the involvement of coexisting silicate phases (Cameron 1975, Henderson 1975, Hamlyn & Keays 1979, Roeder & Campbell 1985).
3. After all the interstitial liquid has crystallized, re-equilibration between chromite and silicate may take place, as temperature changes further at the subsolidus stage (Jackson 1969, Wilson 1982, Hatton & von Gruenewaldt 1985).

Subsolidus re-equilibration between chromite and silicate is proposed to explain the compositional variation in chromite, displayed mainly by divalent cations, between grains related to olivine and those isolated from olivine, based on the following observations:

1. The compositional variation of chromite is closely linked to the distribution of associated silicate minerals, in particular, whether or not chromite grains are in contact with olivine. The position of chromite grains relative to olivine can have been achieved only after settling of all of the cumulus minerals, and therefore the reaction of chromite with magma before or during the course of chromite precipitation can be excluded as the cause for the compositional variation of this chromite.
2. Based on a variety of localities, Hamlyn & Keays (1979) and Henderson & Wood (1981) found that chromite grains trapped in olivine are richer in Fe^{2+} than those trapped in plagioclase. Similarly, Wilson (1982), Hatton & von Gruenewaldt (1985) and Roeder & Campbell (1985) all reported that included chromite is enriched in Fe^{2+} compared with intergrain chromite. In addition, we observe that intergrain chromite can be as rich in Fe^{2+} as included chromite, if it is in contact with olivine (Figs. 3, 4). This relationship clearly suggests that crystal – interstitial liquid reaction, as proposed by Hamlyn & Keays (1979), is not the cause for the observed decrease in Fe^{2+} from included chromite to intergrain chromite. If that process were to operate, then intergrain chromite in partial contact with olivine should have the same composition as chromite entirely in the postcumulus minerals, since both types of intergrain chromite are not enclosed and protected by olivine and would have had an equal opportunity to react with the interstitial liquid.
3. Olivine and pyroxene can accommodate both Fe^{2+} and Mg as major components in their structure, and the

relative amount of the two elements in these minerals will vary principally with temperature. Consequently, a reaction with ferromagnesian silicates could account for the observed variation in divalent cations in the chromite.

4. Compositional variation among the trivalent cations in chromite shows little covariation with the proportions of divalent cations (Figs. 3, 4). Unlike the conclusion reached by Hamlyn & Keays (1979), that melt is needed to balance the redistribution of trivalent cations in chromite, exchange involving divalent cations can be accommodated by reactions involving only chromite and ferromagnesian silicates. This adds further support for our proposal that the reactions took place at the subsolidus stage.

5. Late-stage overgrown "tails" on the intergrain chromite (Fig. 2c) have a similar composition to grains included entirely in olivine. Furthermore, included chromite partly in contact with postcumulus phases has the same composition as included chromite entirely within olivine (Fig. 5). These varieties of chromite would not show similarities in chemistry if their compositions were unchanged from initial crystallization. If subsolidus reactions had not occurred, changing compositions of the melt with time would have ensured that chromite totally included in olivine and isolated from further reactions with melt would be chemically distinct from either late-stage, overgrowth chromite or chromite partially exposed to intercumulus liquid.

Included chromite might have reacted with interstitial liquid by diffusion through the host olivine (Scowen *et al.* 1991). However, it would then be expected that the intergrain chromite would have the same composition as the included chromite, in contrast to the results displayed in Figures 3 to 6. If included chromite reacted with interstitial melt, then the extent of the reaction was low, and its influence would have been obscured by the effects of later reaction at the subsolidus stage.

6. Grain contacts between chromite and silicate minerals are straight and smooth, lack evidence of replacement by either mineral, and are free of new mineral phases. These observations justify the use of equilibrium reactions involving only chromite and associated silicate minerals in discussing subsolidus behavior.

Based on thermodynamic considerations, Irvine (1965) concluded that the distribution coefficient $(Mg/Fe^{2+})_{silicate}/(Mg/Fe^{2+})_{spinel}$ increases with falling temperature. Thus, as the rocks cool, Mg^{2+} will tend to transfer into olivine from chromite, whereas Fe^{2+} will move from olivine to chromite. The subsolidus equilibrium between chromite and olivine has been tested experimentally on natural samples by Roeder *et al.* (1979). In our study, the best example of the re-equilibration reaction is given by chromite in postcumulus plagioclase (Fig. 3), which shows that chromite in partial contact with olivine shifts its composition to a higher $X(Fe^{2+})$, without any obvious change in ratios of trivalent cations. Since no new phases were formed, the components have to be balanced between the olivine and chromite grains in contact. The exchange reaction can be expressed as:

$$\begin{aligned}&Fe_2SiO_4 + 2Mg(Cr,Al,Fe^{3+})_2O_4 = \\ &\quad Mg_2SiO_4 + 2Fe(Cr,Al,Fe^{3+})_2O_4 \qquad (1)\end{aligned}$$

This reaction explains the observed difference in composition between included and intergrain chromites, and between intergrain chromite partly in contact with olivine and that entirely within postcumulus phases.

Olivine, in some instances, may contain Cr (Huebner *et al.* 1976). Microprobe analysis of residual olivine, however, did not reveal any detectable Cr or Al (Yang, unpubl. data, 1992). Based on evidence of subsolidus exchange of cations only at the grain scale (see following discussion), the low contents of trivalent cations noted in olivine preclude any significant change, at the subsolidus stage, in the contents of trivalent cations in chromite.

Al is a major element in plagioclase. However, the structure of plagioclase cannot accommodate any significant amount of either Fe^{3+} or Cr^{3+}. Minor Fe^{2+} and Mg might be present in plagioclase, as recorded in lunar samples (Hafner *et al.* 1971, Schrmann & Hafner 1972, Wenk & Wilde 1973), but the original composition of plagioclase cannot be obtained in this study, due to its extensive alteration. Because of the generally low level of Fe^{3+}, Cr^{3+}, Fe^{2+} and Mg in plagioclase, the potential for cation exchange with chromite is minimal. A significant change in the composition of chromite could only be achieved by cation exchange involving a large volume of plagioclase. This is not indicated by the broad range of $X(Fe^{2+})$ preserved in intergrain chromite, or from the observed relationship between composition and grain size (see following discussion). We conclude, therefore, that the chromite grains located entirely within plagioclase are essentially unaffected by subsolidus re-equilibration, and that their composition may represent a magmatic composition, preserved through the subsolidus stage.

For those grains of chromite wholly within clinopyroxene, no direct evidence is available to judge the possibility of re-equilibration between chromite and clinopyroxene based on Fe^{2+}–Mg exchange. Considering the slight enrichment in Fe^{2+} displayed by included chromite in clinopyroxene compared with chromite in plagioclase from some cumulate lenses, an exchange reaction is likely to be involved, as inferred for the Seiad, California and Finero, Italy complexes (Medaris 1975). The appropriate reaction for Fe^{2+}–Mg exchange between the two minerals can be expressed simply as:

$$\begin{aligned}&Fe_2Si_2O_6 + 2Mg(Cr,Al,Fe^{3+})_2O_4 = \\ &\quad 2Fe(Cr,Al,Fe^{3+})_2O_4 + Mg_2Si_2O_6 \qquad (2)\end{aligned}$$

This reaction appears to be less significant than reaction

(1) involving olivine and has had less impact on subsolidus changes in the composition of the chromite.

Grain-size data acquired for some grains were used to test a possible correlation between composition and grain volume. Chromite grains (both included and intergrain) in physical contact with olivine display a weak trend of decreasing $X(\mathrm{Fe}^{2+})$ with increasing grain-size, similar to observations made on chromite from the Great Dyke by Wilson (1982). Chromite grains wholly in plagioclase do not show any correlation between composition and grain size (Figs. 7, 8).

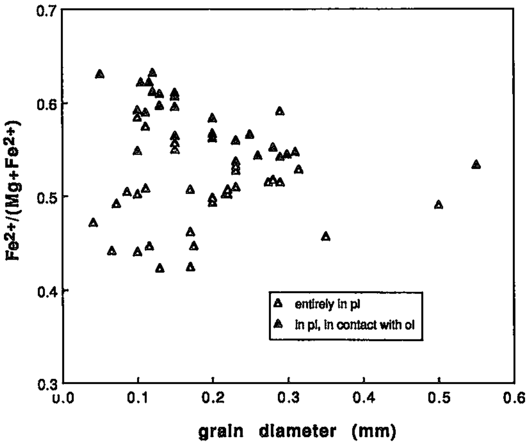

FIG. 7. The composition of chromite in plagioclase, as a function of grain-size distribution.

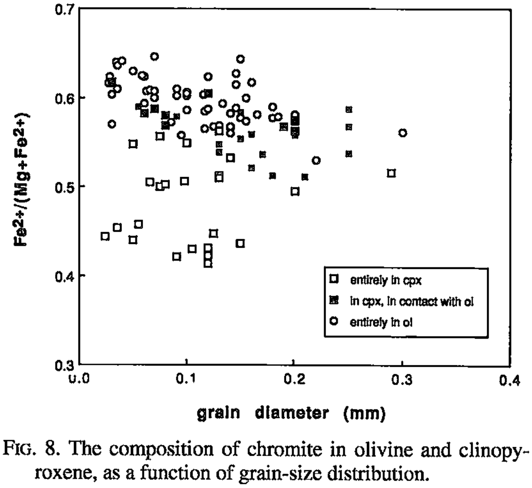

FIG. 8. The composition of chromite in olivine and clinopyroxene, as a function of grain-size distribution.

A similar correlation between chromite composition and grain size was observed in the Panton Sill by Hamlyn & Keays (1979). These authors concluded that chromite continued to react with magma, resulting in a decrease in Fe^{2+} and an enlargement of the grain. If prolonged reaction with magma is the cause for the relationship between composition and grain size, it might be expected that the chromite grains entirely within plagioclase, having a greater chance to react with magma, would exhibit the strongest grain-size effect, in contrast with the included chromite protected in olivine. This would produce a completely opposite picture to the data of Figures 7 and 8. The dependence of chromite compositions on grain size for intergrain chromite in partial contact with olivine, and the lack of correlation between grain size and composition for intergrain chromite entirely within the postcumulus phases, can only result from subsolidus reactions with olivine, rather than a reaction with magma. For chromite partly in contact with olivine, we further infer that the decrease in $X(\mathrm{Fe}^{2+})$ with increasing grain-size results from grain-scale equilibration with olivine, dependent on kinetically driven diffusion-controlled reactions.

Irvine (1967) has pointed out that the change in composition of chromite due to subsolidus reactions with silicate minerals is a function of the relative amounts of chromite and silicate; the more chromite present, the less change occurs in the $\mathrm{Fe}^{2+}/\mathrm{Mg}$ of the chromite. Jackson (1969) found similarly that $X(\mathrm{Mg})$ can be correlated positively with modal percentages of chromite. Hatton & von Gruenewaldt (1985) noted also that the degree of equilibration of chromite with silicate is controlled by the modal abundance of chromite; with a decreasing proportion of chromite, subsolidus diffusion proceeds to lower temperature.

Wilson (1982) suggested that correlations between grain volume and composition applied to local subsystems are controlled by the ratio of surface area to volume of individual grains, whereby the smaller the grain, the lower the blocking temperature for diffusion of divalent cations. Our observations indicate that the subsystem may be represented by each chromite grain plus a portion, or the whole, of an associated grain of a silicate mineral. Cation diffusion between chromite and silicate during re-equilibration is dependent on the kinetics of the exchange reactions and the diffusion path-lengths, which effectively defines the subsystem. The concept of achieving equilibrium in a subsystem comprising adjacent grains is readily verified, as is evident by higher Fe^{2+} contents in intergrain chromite in contact with olivine, compared to neighboring grains from the same postcumulus phase isolated from olivine.

The dimensions of individual subsystems, which are determined by the effective distance of diffusion of the cations, are likely to be consistent. Thus, with an increase in grain size, the volume ratio of chromite to silicate within each subsystem increases (and the ratio of surface area to volume of the chromite grain decreases). Diffusion path-lengths required to achieve cation exchange and re-equilibration between the phases also increase. As a result, the larger a chromite grain becomes, the higher the temperature of effective blocking for the subsolidus reaction and hence, the lower the $X(\mathrm{Fe}^{2+})$ in chromite. The data points in Figures 7 and 8 for intergrain chromite in partial contact with olivine depict this relationship, by defining a general trend of decreasing Fe^{2+} with increasing grain-size of chromite. Regression analysis of the data sets gives correlation coefficients (R^2) for included chromite (0.31), or intergrain chromite in contact with olivine and hosted by either plagioclase (0.49) or clinopyroxene (0.30). Correlation coefficients for chromite grains located entirely in clinopyroxene or plagioclase are 0.06 and 0, respectively. This demonstrates that the composition of chromite associated with olivine correlates much better with grain size than that of chromite isolated from olivine. Scatter in the data, for chromite grains associated with olivine, is acceptable in these diagrams if we consider that grain areas, rather than grain volumes, are being measured.

The lack of correlation between grain size and composition for chromite hosted entirely within clinopyroxene (Fig. 8) fits with the conclusion drawn previously that chromite did not achieve equilibrium during subsolidus reaction with pyroxene. This suggests that the Fe^{2+}–Mg exchange between chromite and olivine is much more effective than that between chromite and clinopyroxene. This difference probably results from the faster rate of diffusion of divalent cations through olivine than that through clinopyroxene, analogous to the greater rate of diffusion for Fe^{2+}–Mg exchange between chromite and olivine compared to the chromite–orthopyroxene pair, established by Wilson (1982), Hatton & von Gruenewaldt (1985) and Roeder & Campbell (1985).

Because of the low rate of diffusion, exchange reactions between chromite and clinopyroxene would have ended at a higher temperature than those between chromite and olivine. It can be anticipated that the rate of diffusion and distance decrease with falling temperature. At higher temperature, relatively slow rates of diffusion of cations between chromite and linopyroxene, compared with the case for chromite–olivine, may be offset by greater effective diffusion-distances, leading to a larger subsystem. This would result in reducing the effect of individual variations in grain size on chromite composition.

Uncertainties remain concerning the actual dimensions of the subsystem, particularly with regard to the proportion of the silicate grain involved in the subsolidus reactions. Olivine grains were almost completely altered, rendering it impossible to identify possible compositional zoning caused by the re-equilibration. Compositional zoning in the relatively fresh clinopyroxene was not encountered.

The composition of chromite grains entirely within plagioclase

The fraction of Fe^{2+} cations in the intergrain chromite hosted entirely in plagioclase varies over a range almost twice as large as that for chromite grains in contact with olivine (Fig. 3). Following from the above discussion, it could be argued that the observed lack of correlation between composition and grain size for chromite grains entirely in plagioclase (Fig. 7) implies that subsolidus re-equilibration had taken place, but in a large subsystem at an overall high temperature. However, this re-equilibration would lead to chromite with a smaller range of $X(\mathrm{Fe}^{2+})$ compared to that in contact with olivine, because the grain-volume effect on chromite composition would be minimized. This suggests that the chromite grains located entirely in plagioclase have retained compositions inherited from above the solidus, either during crystallization from parental magma or modified by reaction with magma. This conclusion also is supported by data presented in the following section dealing with concentrated chromite.

Processes that cause or modify the chemical composition of chromite above the solidus are likely to be more complex than those operating below the solidus. Unlike subsolidus reactions, which are influenced by the local environment, defined by the minerals present, individual grains of cumulus chromite from the same layer could have been generated under quite different T–P–X conditions in diverse portions of the magma. Convection in the magma chamber is likely to play an important role in this process, because of the wide distribution of chromite grains crystallized under a variety of conditions. Thus, grains present even in the same pore space may have experienced quite distinct processes, such that any variation in composition might not be related to a common cause.

Evidence is not available to estimate the effects of solid–liquid interaction between chromite and magma, either in the main body of the magma chamber or within the pile of cumulus crystals. Scowen *et al.* (1991) concluded that re-equilibration between chromite and residual liquid changes the composition of chromite. The wide range in composition for the intergrain chromite and the preservation of distinct differences in composition among adjacent chromite grains, however, seem to rule out large-scale equilibrium between chromite and magma after chromite settling. At the postcumulus stage, equilibrium among silicates, chromite, and interstitial melt in local systems may account for the range in composition of chromite at the mesoscale, but it cannot explain the distinction between adjacent chromite grains in a single pore. Therefore, it appears that differences in composition of intergrain chromite result from crystallization processes and crystal-liquid reactions, which took place before chromite deposition. The compositional variation must have resulted from conditions in the magma chamber, and changes in the physicochemical conditions of the crystallizing magma, even if these processes cannot be quantified.

Temperature, oxygen fugacity and magma composition are the main parameters that control the chemistry of chromite at the magmatic stage. The compositional variation of chromite grains within plagioclase is mainly expressed by $X(Fe^{2+})$, with only minor changes in the fraction of trivalent cations (see Fig. 3). Experimental studies indicate that $X(Fe^{2+})$ of spinel increases with falling temperature (Fisk & Bence 1980, Barnes 1986, Murck & Campbell 1986) owing to olivine fractionation and the increase in K_d^{ol-sp} of Mg–Fe^{2+} (Fisk & Bence 1980),

Irvine (1965) discussed the potential application of $X(Fe^{3+})$ as an indicator of the state of oxidation of a melt. In addition, Murck & Campbell (1986) demonstrated experimentally that $X(Fe^{3+})$ provides the best measurement of a magma's $f(O_2)$. In Figure 3, the $X(Fe^{3+})$ of chromite indicates a constant or slight increase in oxidation state, accompanying an increase in Fe^{2+}. According to the experimental results of Murck & Campbell (1986), a small increase in oxidation state could not cause a corresponding increase in Fe^{2+} as large as that displayed by the chromite, without a much greater accompanying increase in Fe^{3+} content.

On the other hand, if a fall in temperature is the only cause of the decrease in Cr^{3+} content, Al^{3+} would be expected to increase, along with Fe^{2+} (Hill & Roeder 1974, Murck & Campbell 1986). We conclude that the compositional variation results from the effect of more than one parameter. A temperature decrease, coupled with olivine fractionation during crystallization of chromite, appear to be the two most important factors that have led to the wide range in $X(Fe^{2+})$ of chromite. These two factors may have obscured any additional effect due to a change in the melt's $f(O_2)$ on chromite chemistry.

Since Mg/(Mg+Fe^{2+}) of early-formed olivine is much larger than that of basaltic magma (Hamlyn & Keays 1979), fractionation of olivine would effectively lower this ratio in the residual melt, so that chromite would become progressively enriched in Fe^{2+}. Recent experiments by Roeder & Reynolds (1991) clearly demonstrate that $X(Fe^{2+})$ in chromite changes sympathically with that of parental magma.

The large range encountered in $X(Fe^{2+})$ of chromite hosted by plagioclase may not necessarily correspond to a significant change in melt temperature during the crystallization of chromite. The experiments of Arculus *et al.* (1974) reveal that a small change in temperature and liquid composition could generate substantial changes in composition of the associated spinel.

The difference in composition between concentrated and disseminated chromites

The difference in chemistry between disseminated and concentrated chromites is illustrated in Figure 6 for a chromite band in a troctolite host, in which the grains of disseminated chromite in troctolite are only a few millimeters from the interface between the two lithologies. Compositional relationships depicted on Figure 6 are similar to those observed in the Rhum Intrusion (Henderson 1975, Henderson & Wood 1981), Bushveld Complex (Cameron 1977, Hatton & von Gruenewaldt 1985), and the Bird River Sill (Ohnenstetter *et al.* 1986).

Studies elsewhere emphasize the role of subsolidus reactions in generating differences in chemistry between disseminated and concentrated chromites. In discussing the application of the olivine–chromite thermometer, Irvine (1967) favored the use of chromite from chromite-dominant rocks to avoid the influence of subsolidus equilibration. Hatton & von Gruenewaldt (1985) showed that the original composition of disseminated chromite, adjusted for the effects of subsolidus re-equilibration, is similar to that of concentrated chromite.

If the difference shown in Figure 6 was caused by subsolidus reactions under different modal ratios of olivine to chromite, then the grains of concentrated chromite at the interface between chromitite and troctolite would have a composition somewhat different from those wholly within the chromitite band, since they are in direct contact with the olivine-dominant rocks and so occur in an environment of higher olivine/chromite ratio. Analysis of chromite at the margin of the chromitite band indicates no such difference. Instead, a sharp change in composition exists at the interface between the concentrated and disseminated chromites. The compositional differences between concentrated and disseminated chromites appear to result from processes that took place above the solidus, probably during initial crystallization of the chromite, rather than during subsolidus re-equilibration.

The lack of intermediate compositions for chromite grains at the boundary of the chromitite band and the adjacent troctolite also promotes the argument that any subsolidus re-equilibration must be restricted to small-scale subsystems. If the volume of the subsystem was larger than the scale of grains, then high values of $X(Fe^{2+})$ should be observed for those grains of concentrated chromite in proximity to abundant olivine across the chromitite–troctolite contact.

One of the possible mechanisms leading to the formation of the layer of concentrated chromite requires that the liquidus composition of the magma shift into the chromite field from the olivine–chromite cotectic. The shift is likely to result from mixing of the crystallizing magma with a new pulse of magma (Irvine 1977). The repetition of chromitite layers would then represent cyclic pulses in the supply of new magma to the chamber. The mixing could be isothermal, with the new liquid introduced at the same temperature as the residual melt, so as to produce a magma composition below the chromite liquidus surface. In this case, the chromitite layer would crystallize at a similar temperature to the disseminated chromite at the cotectic composition. If the new magma is hotter than the evolved melt, chromitite would form at a temperature higher than the cotectic chromite.

Alternatively, generation of the chromite-rich layers could have resulted from an increase in the state of oxidation of the melt (Ulmer 1969), since the solubility of Cr in melts decreases markedly with a rise in $f(O_2)$ (see the experiments of Murck & Campbell 1986, Roeder & Reynolds 1991). If so, concentrated chromite should indicate a higher $X(Fe^{3+})$ than disseminated chromite. The data of Figure 6 demonstrate, however, that $X(Fe^{3+})$ in concentrated chromite is close to, or slightly lower than, that of disseminated chromite, implying that an increase in oxidation state of the melt is not the main cause for the crystallization of the banded chromitite.

Because concentrated and disseminated forms of chromite probably represent different liquidus assemblages, it is inappropriate to use the chemical differences between the two types of chromite to determine the physicochemical reasons corresponding to the change from olivine–chromite to chromite cumulate. Thus, unlike the arguments proposed for the formation of disseminated chromite, the influence of a change in T, $f(O_2)$ or degree of olivine fractionation in the melt on the crystallization of concentrated chromite cannot be assessed, unless it could be proven that the composition of chromite is independent of the melt's composition.

CONCLUSIONS

1. Subsolidus reactions involving equilibration between chromite and ferromagnesian silicates are an important influence in modifying chromite compositions in olivine–chromite cumulates from the Great Serpentinite Belt, New South Wales. Re-equilibration between chromite and olivine is far more significant in modifying the composition of chromite than the reaction generated between chromite and clinopyroxene. Principal chemical effects in chromite involve the divalent cations and are characterized by an increase in the fraction of Fe^{2+} in chromite in contact with olivine.
2. Subsolidus equilibration operated in grain-scale subsystems comprising a chromite grain plus a part of a surrounding or neighboring grain of a silicate mineral. The degree of equilibration is inversely correlated with grain size; re-equilibration proceeded to lower temperatures in smaller grains of chromite.
3. Chromite grains hosted entirely by plagioclase have been preserved from subsolidus re-equilibration. The large compositional variation encountered in these grains was inherited from the magmatic stage and resulted primarily from a decline in temperature, aided by olivine fractionation at stages above the solidus.
4. The chemical contrast between concentrated chromite in chromitite bands and disseminated chromite is not due to subsolidus reaction, but reflects initial differences in conditions of crystallization for the two types of chromite.

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