The Nature and Origin of the Chrome-Spinel of the Rhum Layered Intrusion

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Abstract. The textural features and mineralogy of chrome-spinel bearing rocks from the layered ultrabasic intrusion of Rhum indicate that *in situ* reaction of chromite with olivine, plagioclase and interstitial liquid has taken place. The compositions of the chrome-spinels vary throughout parts of the intrusion and also in a systematic way across a thin picotite rich seam at a feldspar cumulate—olivine cumulate junction. The origin of the chrome-spinel is discussed in relation to other occurrences of chromite in layered ultrabasic rocks.

Introduction

A study of the origin and nature of chromites or, more generally, chrome-spinels in layered ultrabasic and basic igneous rocks has been made by a number of workers (e.g. Thayer, 1946: van der Walt, 1942: Irvine and Smith, 1969), not only because of the economic implications but because an elucidation of the origin is of importance in an understanding of the formation of these intrusions.

The eastern part of the rhythmically layered intrusion of Rhum was studied by Brown (1956). He showed that the ultrabasic layering originated by rhythmic crystal accumulation from basalt magma and was not a result of successive, sill-like, ultrabasic injections as believed by Harker (1908) and Bailey (1945). Large scale rhythmic units occur, each grading from an olivine-cumulate (peridotite) at the base to a feldspar-cumulate (allivalite) at the top. The mineralogy of the rocks is simple with olivine, clinopyroxene, and plagioclase feldspar as the predominant cumulus minerals. Small amounts of chrome-spinel occur, sparsely distributed throughout most of the intrusion and as thin concentrations at some of the junctions of adjacent, macro-rhythmic units (Brown, 1956). The best development of a chromite seam is at the junction of the feldspar-cumulate forming the top of unit 11 and the olivine-cumulate base of the overlying unit 12. A summary, showing the relationship of the chromite distribution to the layered complex, is given by Wager and Brown (1968, Chapter 10).

There is very little published work on the Rhum chrome-spinel. Harker (1908) gave a brief description and a poor chemical analysis (by Heddle); Brown (1956) described the seam at the base of unit 12 and recorded that the translucency of the spinels within the seam was greater than for those isolated specimens just below the seam. He also noted the presence of some exsolution lamellae of ilmenite in some of the dispersed crystals at the top of unit 11. The absence of any reliable

chemical data and the variation in translucency of an assumed cumulus mineral prompted one of us (PH) to collect material from the intrusion and to investigate the nature of this mineral as part of a general project on the geochemistry of the Rhum layered rocks.

Occurrence and Mineralogy

Chromite Seam. A specimen containing the olivine-cumulate at the base of unit 12, the feldspar-cumulate at the top of unit 11 and the layer of chrome-spinel at the junction was collected from the north-western flank of Hallival (see Fig. 1). The thickness of the seam in this specimen is 1.5 mm. Variations in the thickness were noted in the field and some loose blocks near to the Barkeval eucrite were seen to contain a chromite layer which was markedly undulatory and variable in thickness. In one specimen from a loose block, the seam varies in thickness from 1.5 mm to 5 mm adjacent to a microfault through the seam. In the area of the microfault the associated olivine shows undulatory extinction in thin section.

The seam consists of irregularly shaped chrome-spinel crystals of mean size of 0.2 mm surrounded by large poikilitic plagioclase feldspar crystals (Fig. 3 and Wager and Brown, 1968, Figs. 155–156). The chrome-spinel crystals show only a poor development of synneusis structure. The poikilitic crystals of feldspar show patchy zoning and it is noteworthy that the euhedral plagioclases right at the top of unit 11 also show patchy zoning whereas those immediately beneath them do not.

At the top of the layer there is marked evidence of post-depositional reaction of the unit 12 olivine (Fig. 3). The olivines are strongly corroded against the chrome-spinel but also show some later sub-poikilitic growth developments which extend a short way down into the seam. The extensions to the olivine crystal shown in Fig. 3 are in optical continuity with the cumulus crystal. Observations of thin sections show that there is a change in the translucency of the chrome-spinels across the layer. The top third has chrome-spinels which are more opaque than those in the lower two thirds. Dispersed chrome-spinel crystals either side of the seam (in the olivine-cumulate and in the feldspar-cumulate) are opaque. This distribution confirms the findings of Brown (1956). Examination of a polished mount did not show any exsolution of ilmenite in the chrome-spinel either in the seam or in dispersed crystals up to 1 cm below the seam. An X-ray powder photograph of the chrome-spinel from the seam showed the presence of only a pure spinel phase, and some brief transmission studies with an electron microscope did not reveal any incipient exsolution within the chrome-spinel. The reflected light study revealed the presence of small crystals of chalcopyrite both in the chrome-spinel layer and either side of it in the olivine-cumulate and feldspar-cumulate. The chalcopyrite is interstitial and less than 0.1 mm in size. Only small amounts of it were recorded, there being slightly more in the chrome-spinel layer than in the other rock types. In the seam some chalcopyrite is enclosed by chrome-spinel but the majority is interstitial.

Dispersed Chrome-Spinel. Dispersed crystals occur throughout the intrusion enclosed in olivine crystals and also interstitially to cumulus olivine and plagioclase. The interstitial crystals are often euhedral to subhedral and occur in small clusters.
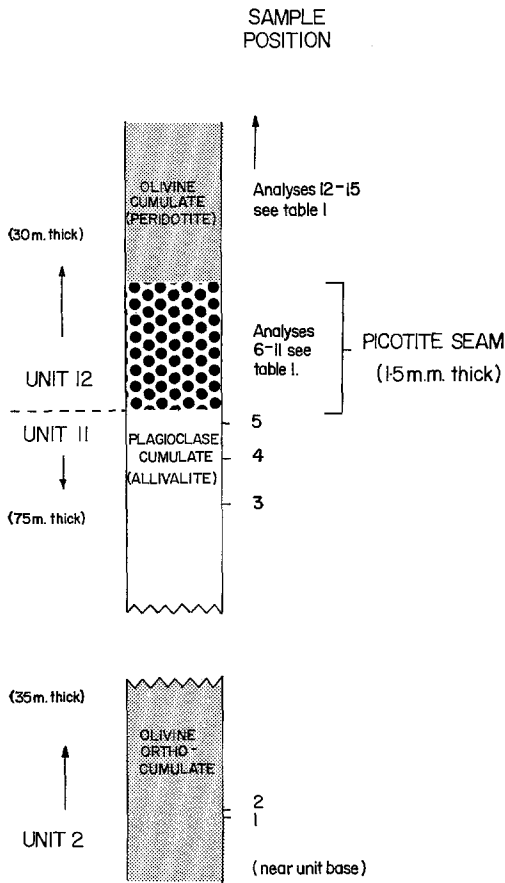
SAMPLE
POSITION

Fig. 1. Position of analysed chrome-
spinels within the layered sequence of
the Rhum intrusion

(30m. thick)

OLIVINE
CUMULATE
(PERIDOTITE)

Analyses 12-15
see table I

UNIT 12

Analyses
6-11 see
table I.

PICOTITE SEAM
(1·5 m.m. thick)

UNIT II

PLAGIOCLASE
CUMULATE
(ALLIVALITE)

5

4

3

(75m. thick)

(35m. thick)

OLIVINE
ORTHO-
CUMULATE

UNIT 2

2
1

(near unit base)

The higher layered units of the intrusion tend to contain more chrome-spinel
than the lower units but there may be a marked variation in modal amount
of chrome-spinel from one rock to another in a unit. For example, in unit 10 much
of the dispersed chromite is to be found right at the top, just below the middle
and at the base of the unit. Other parts of the unit contain virtually no chrome-
spinel.

A significant textural feature associated with the chrome-spinel is that crystals
are frequently located in small embayments in the cumulus olivine crystals, with
a narrow amount of feldspar separating the chrome-spinel from the olivine
(Fig. 4). A number of workers have described this feature in other chromite
deposits (e.g. Thayer, 1946) and Brown (1956, Fig. 40) has described the Rhum
occurrence. However, two points need to be added. Firstly, there is commonly
no feldspar between the chrome-spinel and the olivine at the junction of units 11
and 12 or where the chromite is enclosed within the olivine (Fig. 3), and secondly
that when there is a narrow development of a mineral between the olivine and
chrome-spinel in the Rhum rocks it is nearly always, but not exclusively,
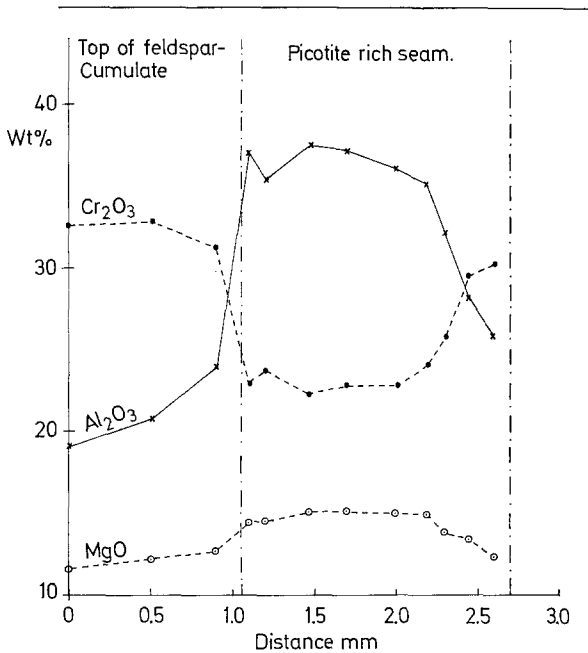plagioclase. Very occasionally it is pyroxene.

Fig. 2. Variation in the Cr_2O_3, MgO and Al_2O_3 contents of homogeneous chrome-spinel crystals across part of the feldspar rich seam and the picotite rich seam at the junction of unit 11 and unit 12. (The diagram is based on thirteen analyses, nine of which are given in the Table. The average of analyses 9 and 10 in the Table has been taken to give the point at 2.44 mm)

Chemistry

The results of the determination, by electron microprobe, of the compositions of chrome-spinel crystals from three units (Fig. 1) within the intrusion are presented in the Table. The FeO and Fe_2O_3 values are calculated from the total iron results on the basis of 24 cations to every 32 oxygen ions and with titanium as Ti^{4+}. Analyses 1 and 2 are of individual grains in unit 2. They are seen to be iron- and chromium-rich spinels of a composition between hercynite and picotite. Each grain is homogeneous and unzoned. The rock which contains these spinels is a feldspar-cumulate with a high amount of pore material from near the base of the unit.

The compositions of the chrome-spinels from the higher units (analyses 3 to 15 in the Table) are in marked contrast to those from unit 2. They are significantly less rich in iron and much richer in aluminium and magnesium. The most remarkable feature shown by these results is the compositional variation across the thin seam (analyses 6 to 11). All these analyses indicate a picotite which is richer in chromium and iron but poorer in magnesium and aluminium contents at the top of the seam compared with the base (Fig. 2). Analyses of chrome-spinels a short distance into the olivine-cumulate above the seam (analyses 12 to 15) show a return to compositions similar to those found at the top of the feldspar-cumulate below the seam. The compositional variation across the picotite rich seam is clearly of importance in any discussion on the origin of the picotite and the layer, and it accounts for the variation in translucency of the crystals seen in transmitted light. Many points on each grain were analysed but they were each found to be of constant composition.

The analyses (12 to 15) of chrome-spinels in the olivine cumulate are of interest. The chromites totally within a cumulus olivine crystal tend to have more chromium and less magnesium and aluminium than those in the small embayments into the olivine crystals described above (Fig. 4). Similarly, the crystals in this part of the unit have more chromium and less magnesium and aluminium than those in the picotite rich seam.

A Mössbauer spectrum of a bulk sample of picotite from the seam indicates that some of the Fe II is in octahedral co-ordination, but a precise figure cannot be given because of the compositional variants included in the bulk sample. However, it is sufficient to show that the picotite is a partially inverse spinel.

Origin

In considering the origin of the Rhum spinels it is necessary to remember the following points:

1. Chrome-spinel grains are found enclosed in cumulus olivine crystals. This is taken to indicate that the spinel crystallized before or at the same time as the olivines.

2. There is virtually no cryptic variation in the cumulus olivines, plagioclase feldspars and clinopyroxenes throughout the 2500 ft. of eastern series layered rocks (Brown, 1956).

3. Layers rich in chrome-spinel only occur at feldspar-cumulate—olivine-cumulate junctions but not all such junctions have spinel-rich layers.

4. Cumulus olivine is frequently embayed or corroded when adjacent to chrome-spinel, which is poikilitically enclosed by plagioclase (or rarely by clino-pyroxene).

5. There are compositional variations in the chrome-spinels from the three units and there is a systematic variation across a spinel-rich layer, 1.5 mm in thickness.

6. The chrome-spinels in the seam are generally larger than those enclosed by cumulus olivine and they show some variation in shape and size (Fig. 3).
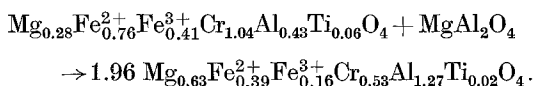
7. If the Rhum parental basalt magma contained a chromium concentration of about 300 ppm. (Brown, 1956, Table 8) then chrome-spinels containing about 33% Cr_2O_3 are enriched in chromium compared with the magma by a factor of about 800.

We suggest the following sequence of events to account for the observations presented in this paper:

a) During the formation of each major unit, chromite was the first cumulus phase to crystallize. Some sank to the floor of the magma chamber to form a chromite rich layer on top of the feldspar-cumulate of the previous unit. Some remained in suspension and sank slowly through the magma. In the formation of some units a spinel rich layer was not formed—all the chromite remained in suspension.

b) Olivine was the next cumulus phase to crystallize and it sank or was carried to the chamber floor to form a layer on top of the chromite and interstitial liquid. The crystallization of chromite and olivine before feldspar and pyroxene in the Rhum intrusion is suggested by Wager and Brown (1968, p. 292), who also discuss

causes for the rhythmic deposition. Thus we have a situation where olivine and chromite crystals at a temperature probably close to that for the melting of the feldspar are resting on a feldspar-rich layer. It is not suggested that there was sufficient heat to melt some of the pre-existing plagioclase feldspar but that the disequilibrium conditions were such that reaction took place between the plagioclase, chromite, olivine and some interstitial liquid. For descriptive purposes only, the reaction can be visualised as taking place between olivine and plagioclase to give spinel and a liquid. The spinel enters into solid solution with the pre-existing chromite to give a picotite. It is not possible to define a precise equation for this reaction but the feasibility of the concept may be checked by consideration of the following:

$$Mg_{0.28}Fe^{2+}_{0.76}Fe^{3+}_{0.41}Cr_{1.04}Al_{0.43}Ti_{0.06}O_4 + MgAl_2O_4$$

$$\rightarrow 1.96\ Mg_{0.63}Fe^{2+}_{0.39}Fe^{3+}_{0.16}Cr_{0.53}Al_{1.27}Ti_{0.02}O_4.$$

The chromite on the left hand side is that of analysis 13 and the picotite on the right is analysis 6. The equation shows a tolerable balance when a mole of chromite and about one mole of spinel react to give 1.96 moles of picotite but the fact that it is not entirely self consistent (especially with respect to Fe^{3+} and Ti) shows that such an equation is an oversimplification.

The reaction of olivine and plagioclase to give spinel was suggested by Bowen (1928) but as pointed out by him, and as seen from phase relationships in the system diopside-forsterite-anorthite (Osborn and Tait, 1952), spinel is unstable as the temperature falls and in reaching equilibrium would react back to form plagioclase and olivine. However, this is no serious problem if the spinel (i.e. $MgAl_2O_4$) enters into solid solution with a chromite. It is possible that Cr^{3+} ions are forced to occupy some tetrahedral co-ordination sites in a basaltic melt, as has been suggested for some other transition element ions (Burns and Fyfe, 1964, 1966; c.f. Seward, 1971). Cr^{3+} occupies octahedral sites in a chrome-spinel and it is well known that in relation to tetrahedral co-ordination, Cr^{3+} has a strong crystal-field octahedral site preference energy in oxide structures (37.6 kcal mole^{-1}, Dunitz and Orgel, 1957). Hence the presence of Cr^{3+} in a spinel (*sensu lato*) should significantly stabilize the mineral in relation to spinel (*sensu stricto*) as magnesium and aluminium have no crystal-field octahedral site preference energy (see Navrotsky and Kleppa, 1967).

It is seen from the analyses that the picotite from the seam in relation to the chromite either side of the seam, is greatly enriched in aluminium but only slightly enriched in magnesium. If the chromite enclosed within the olivine crystals at this level approximates in composition to the initial cumulus chromite then it appears that for every mole of picotite formed, plagioclase and chromite are the predominant reactants. Hence we suggest that the reaction has proceeded further at the base of the seam than at the top. Support for this suggestion comes from the fact that the chromite in the feldspar layer of unit 11 is slightly poorer in chromium and richer in aluminium than the chromite in the unit 12 olivine-cumulate. In the former the reaction would be of the kind plagioclase + interstitial liquid + chromite → picotite + liquid, whilst in the latter it would be olivine + interstitial liquid + chromite → picotite + liquid. The reduced availabil-

Table 1. *Chemical composition of Rhum chrome–spinels*

	1	2	3	4	5	6	7	8
Height[a]	—	—	0	0.5	0.9	1.1	2.2	2.3
Cr_2O_3	29.02	24.91	32.68	32.88	31.27	22.98	24.13	25.82
MgO	5.88	4.54	11.54	12.10	12.58	14.45	14.88	13.84
FeO	26.70	29.10	18.92	18.61	17.07	15.89	15.54	15.98
Fe_2O_3	26.96	31.25	13.79	12.12	12.01	7.40	9.05	10.19
Al_2O_3	7.60	4.88	19.13	20.66	23.86	37.06	35.16	32.08
TiO_2	4.06	5.07	2.54	2.71	1.47	0.81	0.80	0.80
MnO	1.11	0.99	0.20	0.26	0.19	0.12	0.14	0.18
NiO	0.31	0.32	[b]	[b]	[b]	[b]	[b]	[b]
Total	101.64	101.06	98.80	99.34	98.45	98.71	99.70	98.89

Number of ions on basis of 4 Oxygens

Cr	0.78	0.70	0.83	0.82	0.77	0.53	0.56	0.61
Mg	0.30	0.24	0.55	0.57	0.58	0.63	0.64	0.61
Fe^{2+}	0.76	0.86	0.51	0.49	0.44	0.39	0.38	0.40
Fe^{3+}	0.70	0.83	0.33	0.29	0.28	0.16	0.20	0.23
Al	0.31	0.20	0.72	0.77	0.88	1.27	1.21	1.13
Ti	0.10	0.14	0.06	0.07	0.04	0.02	0.02	0.02

	9	10	11	12[c]	13[d]	14[c]	15[d]
Height[a]	2.44	2.44	2.6	8.9	10.9	11.5	11.8
Cr_2O_3	27.71	29.82	30.27	37.03	37.91	35.14	36.62
MgO	13.65	13.10	12.28	7.16	5.40	9.87	5.58
FeO	16.59	16.84	17.89	24.44	26.18	20.17	25.95
Fe_2O_3	9.92	11.78	12.50	13.76	15.77	12.71	16.18
Al_2O_3	31.85	27.35	25.87	12.91	10.51	18.17	12.16
TiO_2	0.86	0.95	1.02	2.65	2.10	1.64	1.78
MnO	0.18	0.20	0.22	0.35	0.40	0.31	0.44
NiO	[b]	[b]	[b]	[b]	[b]	[b]	[b]
Total	100.76	100.04	100.05	98.30	98.27	98.01	98.71

Number of ions on basis of 4 Oxygens

Cr	0.63	0.71	0.73	1.00	1.04	0.91	1.00
Mg	0.60	0.59	0.56	0.36	0.28	0.48	0.29
Fe^{2+}	0.41	0.43	0.46	0.70	0.76	0.55	0.75
Fe^{3+}	0.22	0.27	0.29	0.35	0.41	0.31	0.42
Al	1.11	0.98	0.93	0.52	0.43	0.70	0.49
Ti	0.02	0.02	0.02	0.07	0.06	0.04	0.05

[a] Height in mm above arbitrary base at analysis position 3 (see Fig. 2).
[b] NiO content approximately 0.2% (not included in total).
[c] Analysed chromite in embayment in olivine.
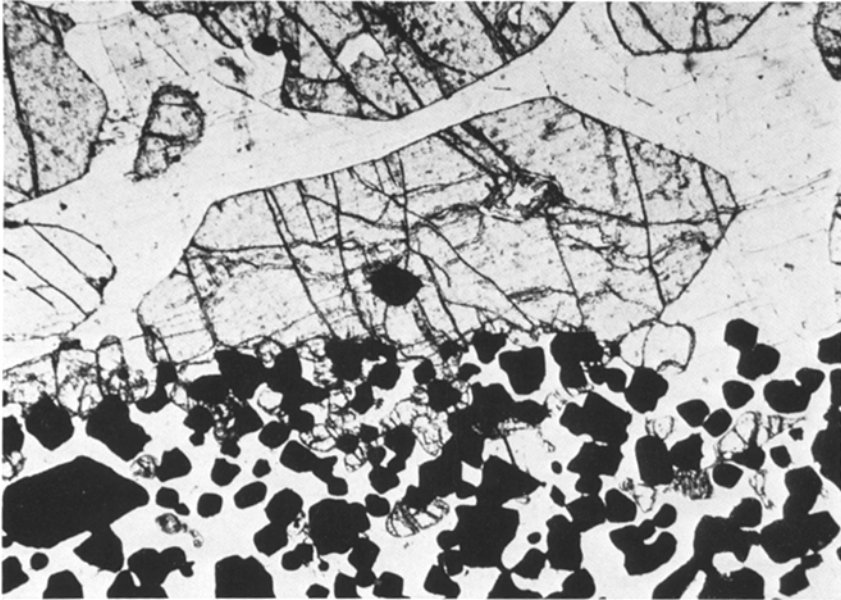[d] Analysed chromite within olivine crystal.

Fig. 3. Cumulus olivine crystal showing corrosion against a picotite-rich layer. Some adcumulus extension to the olivine has occurred. Picotite and olivine crystals are poikilitically enclosed by plagioclase feldspar. Plane polarised light. (Base of unit 12.) Mag. ×58
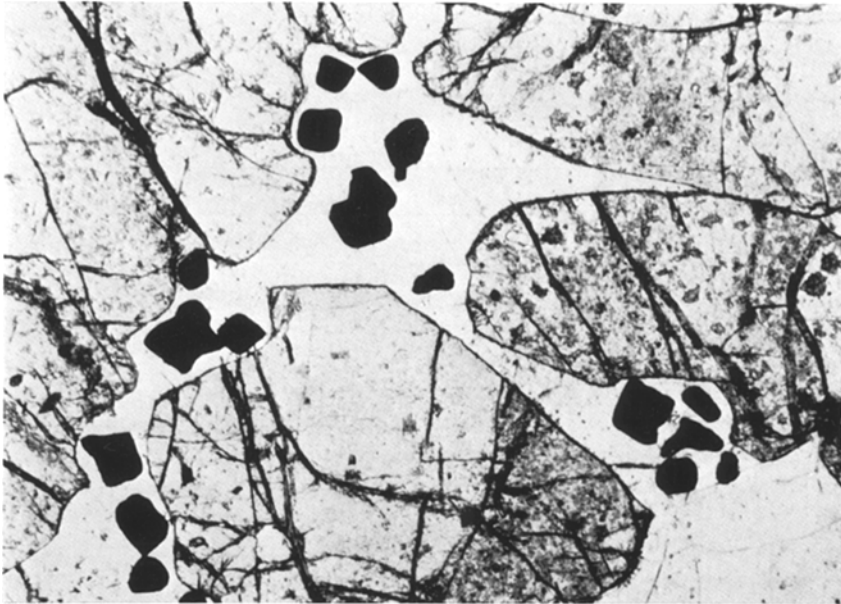

Fig. 4. Cumulus olivine with small embayments containing chromite poikilitically enclosed by feldspar. (Base of unit 12.) Mag. ×58

ity of aluminium in the olivine-rich environment would prevent the reaction from proceeding very far.

c) Reaction at the feldspar-olivine junction of the two units was followed by a small amount of growth of the olivines and of poikilitic plagioclase into the pore spaces.

d) Finally, crystallization of a small amount of trapped liquid occurred which is manifest as zoning of the plagioclases at the top of unit 11 and in the picotite-rich layer.

The extremely iron rich, titanium bearing, chrome-spinels of unit 2 are interpreted as resulting from extended reaction with a relatively large amount of trapped liquid (the rock is an orthocumulate) over a wide temperature range. Zoning of the silicates and the presence of orthopyroxene and apatite in the cumulate led Brown (1956) to this conclusion, but he did not have the evidence of Ti enrichment from the trapped liquid.

Discussion

Some important contributions to our understanding of the nature and origin of chromite deposits in layered rocks by Jackson (1969), Irvine and Smith (1969), Ulmer (1969), Cameron and Desborough (1969), and others have appeared in a recent Symposium Volume on Magmatic Ore Deposits. These contributions show that there are some similarities in chromite deposits from different layered intrusions and some of these are shown by the Rhum chrome-spinels. Irvine and Smith (1969) describe the chromite in the Muskox intrusion and show that, as in the Rhum intrusion, much of it is disseminated throughout the olivine-rich rocks but there is the development of two thin layers near the top of the ultramafic section. Within these layers the chromite is surrounded by poikilitic plagioclase and pyroxene and there is extensively replaced olivine. Furthermore, they attribute the origin of some chromite with polygonal crystal boundaries to recrystallization after deposition. Jackson (1963, 1969) has shown that the composition of chromite varies along the strike of the single chromite zones of the Stillwater Complex and also records, as have other workers on other intrusions, that the composition of the chromite is sometimes a function of the host rock type and may vary over short stratigraphical distances in a layered complex. Jackson (1961) has also described textures in Stillwater rocks which show the secondary enlargement of chromite crystals. Cameron and Desborough (1969) note that whilst the composition of chromite within a chromite rich sequence of the Eastern Bushveld Complex is unrelated to the type of silicate minerals in the associated rock types, it is related to the ratio of the amount of chromite to the amount of silicate at any given horizon.

It is clear from the above discussion that *in situ* reaction of cumulus chromite crystals with adjacent silicates and with interstitial liquid, is thought to be of common occurrence in many ultrabasic and basic layered igneous rocks.

The suggested post-depositional reaction of the cumulus chromite of Rhum to give the picotite-rich layer could be of significance in the discussion of the feasibility of the process of adcumulus growth or overgrowth of *in situ* chromite crystals. Cameron and Desborough (1969) state that artificial chromitites produced

by settling chromite grains through a liquid contain 55 to 70 percent of chromite, yet many natural chromitites contain up to 97 percent chromite. The introduction of overgrowth, by a diffusion process, of *in situ* chromite crystals raises new difficulties (Jackson, 1961; McDonald, 1965; Cameron, 1969) especially in that overgrowth would require diffusion of chromium ions over considerable distances, because chromite contains about 800 times more chromium than the contemporary magma. To overcome this problem, McDonald (1965, 1967) suggested that an immiscible chromite-rich liquid could account for high chromite percentages in chromitites and also account for the presence of rounded silicate inclusions in some of the Bushveld chromites, but pertinent objections to this suggestion have been made by Jackson (1966). The described mechanism for the Rhum picotite formation may be partly applicable to these cases. An estimate of the increase in volume through reaction of the *in situ* cumulus deposit can be made for the Rhum picotite layer on the basis of change in chemical composition, providing one may assume no significant loss of chromium during reaction. If the chromite within the olivine crystals of unit 12 approximates in composition to that of the initial cumulus chromite then the volume of the chrome-spinel in the seam increased by about 50% through post-depositional reaction. Furthermore, it is suggested that post-depositional reaction and enlargement of this kind could produce chromite crystals with inclusions similar to those described by McDonald (1965, 1967) from the Bushveld intrusion.

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