Electron-probe microanalysis of coronas in a troctolite from Sulitjelma, Norway

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Summary. The results of the electron-probe microanalysis of the three minerals from orthopyroxene–amphibole–spinel coronas between olivine and plagioclase in a troctolite from Sulitjelma, Norway, are presented. They are shown to be incompatible with a late-magmatic origin for the coronas, but compatible with an origin by diffusion of material in solution in an aqueous medium across the olivine–plagioclase interface. This theory is discussed on a quantitive basis in the light of the microanalytical data.

REACTION zones, or 'coronas', between olivine and plagioclase are often found in coarse-grained basic or ultrabasic igneous rocks. The reaction zones may consist of a variety of minerals but commonly two zones are present: a zone of orthopyroxene next to the olivine and a zone of an intergrowth of amphibole and spinel next to the plagioclase (Brögger, 1934; Shand, 1945). This paper describes an electron-microprobe study of such zones from a troctolite in the narrow marginal facies of a layered basic igneous pluton at Sulitjelma, northern Norway. The field relationships and petrology of this pluton have been described elsewhere (Vogt, 1927; Mason, 1967) and it is sufficient to state here that the dominant rock-type is olivine-gabbro, and that coronas surrounding olivine are common, especially near the margins of the intrusion. The pluton has undergone partial regional metamorphism to amphibolite, but there is good textural evidence that this occurred after the formation of the coronas.

Fig. 1 shows a photomicrograph of a corona from the specimen that was studied with the electron-probe microanalyser. There are three distinct reaction zones: that nearest the olivine is composed of orthopyroxene prisms with their *c*-axes approximately perpendicular to the interface with the olivine; the middle zone consists of pale green amphibole, and the zone next to the plagioclase is a vermicular intergrowth of this amphibole with colourless spinel. This intergrowth will be referred

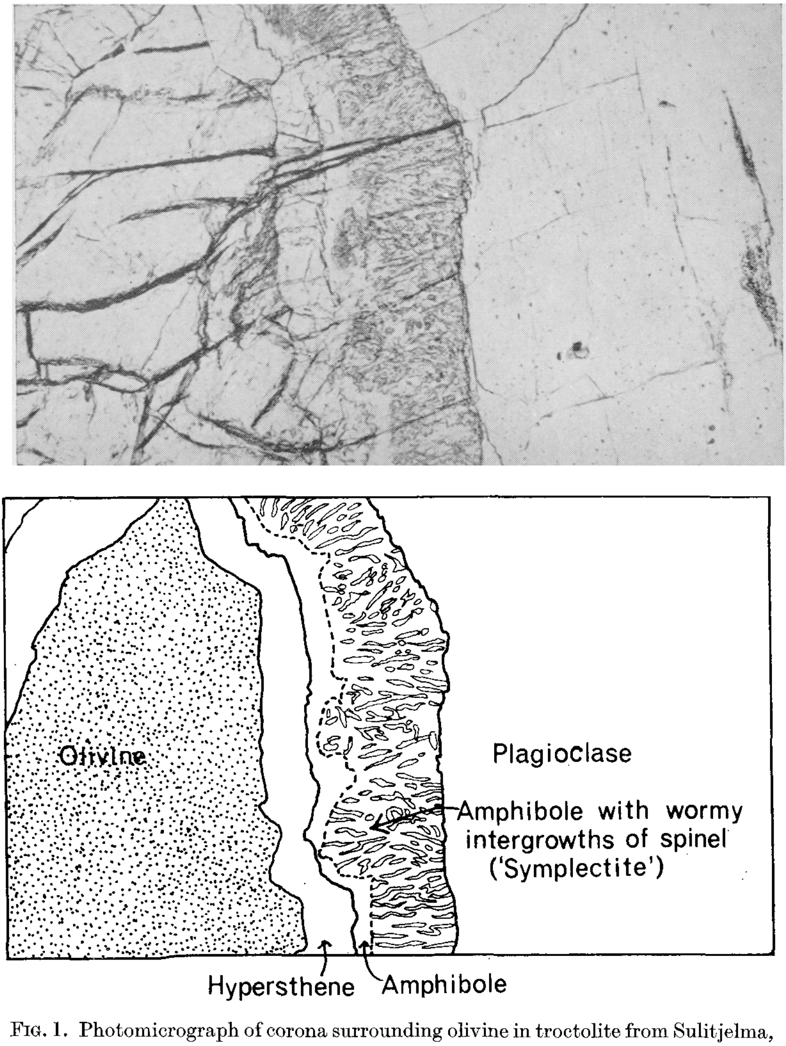

FIG. 1. Photomicrograph of corona surrounding olivine in troctolite from Sulitjelma, Norway (Harker no. 96985). × 250.

to in this paper as an amphibole–spinel symplectite (Sederholm, 1916). There is no optical discontinuity between the amphibole of the symplectite and that of the middle zone.

Many petrologists have advanced theories of the origin of coronas. Reviews of the literature on the subject have been given by Sederholm

(1916), Brögger (1934), and Shand (1945). The theories fall into three broad categories: reactions between early crystals and late magma (Harker, 1909; Herz, 1951; Weedon, 1965); diffusive reactions involving transport of material in solution in an aqueous medium across the interface between olivine and plagioclase (Vogt, 1921; Brögger, 1934; Murthy, 1958); and diffusive reactions involving the introduction and removal of material in solution along the plagioclase–olivine interface (Sederholm, 1916; Shand, 1945). Nearly all writers on the subject are careful to point out that coronas can arise by a variety of mechanisms, and that their conclusions apply to the particular coronites they have studied.

The electron-probe microanalyser is ideally suited to the analysis of fine-grained mineral zones and intergrowths, although even at the best resolutions the spinel grains in the symplectite are often too small to be analysed. One coarser than average specimen from Sulitjelma has been analysed (no. 96985 in the Harker slide collection catalogue in the Department of Mineralogy and Petrology, Cambridge University). The conclusions presented later concerning the origin of coronas apply only to this specimen, but it seems fair to extrapolate them to other orthopyroxene–amphibole–spinel coronas from Caledonian basic intrusions because of the marked similarity between these coronas and the one studied. Coronas have been reported from other parts of northern Norway in gabbros from Seiland (Barth, 1952), Sørøy (Sturt, pers. comm., 1966), Andopen (Vogt, 1927) and Raisuoddar-Haldde (Hausen, 1942). They are also found in the 'Younger Gabbros' of the NE. Highlands of Scotland at Huntly (Read, 1923), Haddo House (Read, 1935), Insch (Read *et al.*, 1961), and Belhelvie (Stewart, 1946), while examples have been described from similar environments in many other parts of the world.

Apparatus and accuracy of results. The analyses were performed on an electron-probe microanalyser in the Department of Mineralogy and Petrology, Cambridge University. This instrument has been developed by J. V. P. Long, and a recent account of its design and operation has been given by Reed (1964). According to Reed, the absolute accuracy of the instrument under optimum conditions is about ±1 % of the concentration of an element for concentrations over about 10 %. The accuracy of the analyses here is less than this: about ±2 % in the orthopyroxene and amphibole analyses and ±5 % in the spinel analyses, except in the case of magnesium, where the errors are about twice these magnitudes.

TABLE I. Analytical data on the troctolite, its coronas, and the constituent minerals

Analyses of plagioclase, olivine, and coronas

	Olivine	Orthopyroxene	Symplectite	Plagioclase
SiO_2	39·4	55·5	40·0	50·7
Al_2O_3	0·0	1·4	24·6	31·9
FeO*	16·7	12·0	6·5	0·2
MgO	43·4	33·4	18·4	—
CaO	trace	0·2	9·8	14·1
MgO : (MgO+FeO)	0·722	0·736	0·739	—
Molecular composition	$Fo_{82}Fa_{18}$	$En_{83}Ofs_{16·7}Wo_{0·3}$	—	$Ab_{33}An_{67}$

The analyses of olivine and plagioclase are based on complete wet chemical analyses of separated powders. The orthopyroxene analysis is the average of nine electron probe microanalyses. See the text for the derivation of the symplectite analysis.

Electron probe microanalyses of phases in the symplectite

	Amphibole (average of 6 analyses)	Spinel (average of 2 analyses)
SiO_2	45·5	2·8
Al_2O_3	18·0	68·2
FeO	5·4	14·1
MgO	18·9	15·4
CaO	11·1	0·5
Total	98·9	101·0

Bulk rock analysis

SiO_2	46·51
TiO_2	0·22
Al_2O_3	20·39
Fe_2O_3	0·18
FeO	5·45
MnO	0·11
MgO	14·35
CaO	10·62
Na_2O	1·97
K_2O	0·14
H_2O+	0·58
H_2O-	0·09
Total	100·61

Modal analysis (vol. %)

Plagioclase	59·4
Symplectite	14·9
Olivine	14·5
Orthopyroxene	9·5
Clinopyroxene	1·8

* Total iron as FeO.

Results

Table I gives the results of the electron-probe microanalyses of the coronas; all the analyses were made around a single olivine grain. Table I also gives the result of a wet chemical analysis of the bulk rock and of separated olivine and plagioclase. The mode of the troctolite is also given, based on a count of 2000 points made on a single thin section. All the orthopyroxene in the mode is from the coronas, and the amphibole and amphibole–spinel zones are included together as 'symplectite'. Thus corona minerals occupy 24·4 % of the rock.

Orthopyroxene. Nine analyses of orthopyroxene were made. In order to find out whether there was any zoning within the orthopyroxene zone two analyses were made in the same sector of the zone, but at different distances from the contact with the olivine. All the analyses gave identical results, within the limits of experimental error, and yield a composition of $En_{83}Of_{16\cdot7}Wo_{0\cdot3}$.

Amphibole. The six analyses of amphibole are not complete, as the instrument used cannot detect H, O, and Na. The proportions of the five elements analysed are consistent with those of a rather aluminous hornblende. Unit-cell contents on a basis of 24 (O, OH) have been calculated according to this assumption by the method described by Deer *et al.* (1966). Na and OH contents and an $Fe^{2+}:Fe^{3+}$ ratio similar to those of a chemically analysed hornblende from the Sulitjelma gabbro complex (Mason, 1966) were used in the correction of the results.

Spinel. In order to increase the resolving power of the instrument, the current in the electron beam was decreased to $\frac{1}{5}$ its usual value for the spinel analyses. This increased the effective power of the electron lenses, but also reduced the number of counts obtained for any element to $\frac{1}{5}$ its usual value, which in turn caused the reduction in accuracy mentioned earlier.

The analyses of the grains intergrown with the amphibole of the symplectite confirm that they are of spinel. The molecular proportions of Si and Al show a slight excess above 16 in a unit cell of 32 oxygens. This is probably due to lack of resolution by the instrument causing some emission of Si radiation from the surrounding amphibole. The deficiency in the Mg and Fe is probably due to the greater inaccuracy of the Mg analysis.

In order to compare the compositions of the spinel and amphibole phases in the symplectite in a qualitative way, scanning pictures of a small area were taken for Mg-$K\alpha$, Al-$K\alpha$, Si-$K\alpha$, Ca-$K\alpha$, and Fe-$K\alpha$ radiations (fig. 2). These pictures show that the spinel phase contains virtually no Si or Ca, while the amphibole has far less Al and Fe than the spinel.

The bulk composition of the symplectite was calculated from the scanning pictures in fig. 2. This gave SiO_2 40·0, Al_2O_3 24·6, total iron as FeO 6·5, MgO 18·4, CaO 9·8, total 99·3 %. This estimate was made by the following method: The Al-$K\alpha$ and Ca-$K\alpha$ pictures that gave the best 'positive' and 'negative' images of the spinel were traced, and a composite tracing based on these was used to estimate the proportion of the surface of the specimen that was composed of spinel. The Ca-$K\alpha$

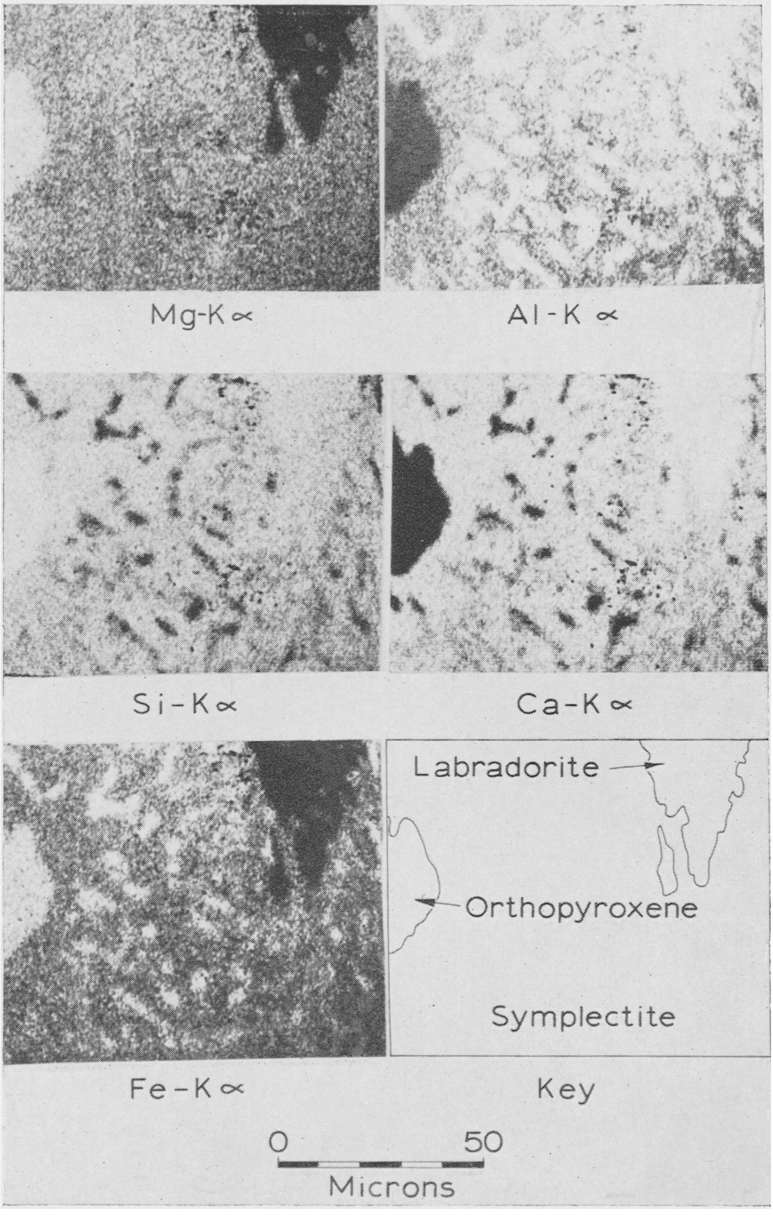

FIG. 2. X-ray scanning pictures of symplectite for various elements.

picture suggested a smaller area of the spinel than the composite tracing, and the Al-$K\alpha$ picture a larger area. The tracing was point-counted at 1 mm intervals (a total of 2941 points being counted) and the weight percentages of the analysed components in the symplectite calculated from the compositions of amphibole and spinel given in table I and data on the specific gravity of these minerals from Deer *et al.* (1963). The amphibole was assumed to be a hornblende.

Zoning of plagioclase and olivine. Optical determinations of the compositions of the cores and margins of plagioclase crystals by the Carlsbad–albite extinction method revealed only slight zoning (less than $\pm 2\ \%$ An). This zoning is usually normal, but occasionally irregular or even reversed.

An examination of the olivine with the electron-probe failed to produce unambiguous evidence of zoning. Some crystals showed no zoning at all. Others showed a decrease of about 4 % in the apparent concentrations of iron and magnesium near the margins. Traverses performed on several occasions showed this trend, which was too great to be due to random counting variation. However, true normal zoning would produce an increase in concentration of iron near the margins and a decrease in magnesium. Attempts to discover any zoning of Al, Ca, and Ni were unsuccessful. These elements are present in uniform trace quantities throughout the olivine. It seems probable that the decrease observed in both iron and magnesium is due to some instrumental effect; possibly a slight slope in the surface of the olivine near the edges of the grains.

Thus it appears that primary zoning of plagioclase and olivine is sufficiently small to be neglected in calculations concerning the mode of formation of the coronas.

Interpretation of the results. Table I shows the results of the electron-probe microanalyses of the coronas, and also of wet chemical analyses of separated olivine and plagioclase from the troctolite. The electron-probe analyses have been corrected for counter dead-time, background radiation, atomic number, and absorption effects by methods described by Long and Reed (1964). The fluorescence corrections are negligible.

The values of the $MgO:(MgO+FeO)$ ratios make a late magmatic origin for the coronas unlikely. There is abundant evidence that minerals that crystallized from late magmatic fluids show a lower magnesium to iron ratio than those that crystallized earlier, but the two corona reaction zones show much the same ratios as the earlier-crystallized olivine. The present writer would also consider theories of reaction between early crystals and late magma implausible on textural grounds, for as Sederholm, Shand, and others have pointed out, it is possible to recognize reaction borders produced from intercumulus magmatic liquids, and these are different from coronas.

It is more difficult to decide whether diffusive reactions are involved. In the following section a diffusive-type hypothesis will be defined and applied to the chemical compositions of the reaction zones of the coronas but, as Shand noted, the greatest difficulties in theories of this type arise over the sodium content of the amphibole, which has not been determined in this study. The hypothesis outlined here is similar to the theories of Vogt (1921) and Brögger (1934), as developed by Murthy (1958), who emphasized the role of an aqueous fluid medium in ionic transfer.

The assumptions made in this hypothesis are: That the present symplectite–orthopyroxene interface represents the original plagioclase–olivine interface; this is considered reasonable on textural grounds (fig. 1); it follows that orthopyroxene formed from olivine, and symplectite from plagioclase. That the mobile elements were magnesium and iron, and to a much smaller extent aluminium and calcium; silicon was passive. And that the replacement of olivine by orthopyroxene and plagioclase by symplectite took place at constant volume.

In accordance with the first two assumptions the composition that arises if FeO and MgO are added to the plagioclase analysis until the weight percentage of silica drops to that in the symplectite (40·0 %) may be calculated. The MgO:FeO ratio is kept at the same value as in the olivine. This gives the result: SiO_2 40·0, Al_2O_3 25·1, FeO 5·8, MgO 15·4, CaO 11·1.

On the assumption that this composition change took place at constant volume an increase in the specific gravity of the symplectite over that of the plagioclase by 21·2 % is indicated. The assumed specific gravity of the symplectite that was used in the calculation of the bulk composition of the symplectite is 3·20. The composition of the plagioclase implies a specific gravity of 2·72 (Deer *et al.*, 1963). These figures give an increase of specific gravity of 17·6 %, and thus the hypothesis accounts for the change from plagioclase to symplectite.

However, difficulties arise over the alteration of olivine to orthopyroxene. It is possible to obtain a close approximation to the composition of the orthopyroxene by subtracting FeO and MgO in the appropriate ratio from the olivine composition (Murthy, 1958). But this only demonstrates that the MgO:(MgO+FeO) ratios are similar, as both phases consist almost entirely of the three components FeO, MgO, and SiO_2. The assumption of equal volume replacement meets severe difficulties because orthopyroxene and olivine of these compositions have almost the same specific gravities, and yet the postulated mobility of iron and magnesium means a reduction in the mass of the olivine on its conversion to orthopyroxene. In the specimen under study, orthopyroxene constitutes 9·5 % of the rock, which implies a reduction in the total volume of the rock of some 3·8 % when the coronas formed. There is no textural evidence for such a reduction.

If the assumption of equal volume replacement is retained, further difficulties are encountered over the chemistry of the coronas. It becomes necessary to assume that silicon and aluminium were mobile and not only diffused across the orthopyroxene–symplectite interface, but also across the symplectite–plagioclase and orthopyroxene–olivine interfaces. This seems unlikely to have occurred in view of the lack of zoning in the olivine and the slight zoning of the plagioclase.

In spite of the volume difficulties, the writer considers this simple diffusion model to be a plausible one for the formation of the coronas, and, by implication, of similar coronas from olivine-bearing basic igneous rocks. It is likely that the diffusion took place through an aqueous fluid that permeated the rock along grain boundaries, and penetrated the olivine and plagioclase for a small distance on either side of the interface.

Conclusion. The theory proposed for the origin of the coronas studied may be summarized as follows: They formed by reaction between olivine and plagioclase at temperatures well below their melting points. A simple process of two-way diffusion across the original interface between olivine and plagioclase, with iron and magnesium diffusing into the plagioclase, and a little aluminium and calcium diffusing into the olivine, explains the chemical composition of the coronas quite well, but creates volume difficulties. The formation of amphibole from plagioclase indicates that water must have been introduced when the coronas formed; the more extensive development of coronas near the contacts of the gabbro complex suggests that this water may be derived from the country rocks; it was probably the transporting medium for ions across the plagioclase–olivine interface.

In describing the coronas as the products of diffusive reaction, it is not intended to imply that they are necessarily the products of metamorphism, in the sense of a reheating of the rocks of the gabbro complex after initial cooling from magmatic temperature. They may be due to reaction occurring during relatively slow cooling from initial high-temperature conditions after consolidation. The Sulitjelma gabbro complex has had a complex history of deformation and recrystallization, and the distinction between the effects of primary cooling and those of regional metamorphism is not easy to make. In any case this distinction can only be made on the basis of an extensive field and petrographic account of the corona-bearing rocks, which would be inappropriate here.

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