Disequilibrium in the Ross of Mull Contact Metamorphic Aureole, Scotland: a Consequence of Polymetamorphism

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The Ross of Mull pluton consists of granites and granodiorites and intrudes sediments previously metamorphosed at amphibolite facies. The high grade and coarse grain size of the protolith is responsible for a high degree of disequilibrium in many parts of the aureole and for some unusual textures. A band of metapelite contained coarse garnet, biotite and kyanite prior to intrusion, and developed a sequence of textures towards the pluton. In Zone I, garnet is rimmed by cordierite and new biotite. In Zone II, coarse kyanite grains are partly replaced by andalusite, indicating incomplete reaction. Coronas of cordierite + muscovite around kyanite are due to reaction with biotite. In the higher-grade parts of this zone there is complete replacement of kyanite and/or andalusite by muscovite and cordierite. Cordierite chemistry indicates that in Zone II the stable AFM assemblage (not attained) would have been cordierite + biotite + muscovite, without andalusite. The observed andalusite is therefore metastable. Garnet is unstable in Zone II, with regional garnets breaking down to cordierite, new biotite and plagioclase. In Zone III this breakdown is well advanced, and this zone marks the appearance of fibrolite and K-feldspar in the groundmass as a result of muscovite breakdown. Zone IV shows garnet with cordierite, biotite, sillimanite, K-feldspar and quartz. Some garnets are armoured by cordierite and are inferred to be relics. Others are euhedral with Mn-rich cores. For these, the reaction biotite + sillimanite + quartz → garnet + cordierite + K-feldspar + melt is inferred. Using a petrogenetic grid based on the work of Pattison and Harte, pressure is estimated at 3-2 kbar, and temperature at the Zone II–III boundary at 650°C and in Zone IV at least 750°C.

KEY WORDS: contact metamorphism; disequilibrium

INTRODUCTION

Contact metamorphic aureoles provide the best-constrained settings in which to interpret metamorphic textures and assemblages (Kerrick, 1991). In principle, knowledge of the shape and size of the intrusion, together with the temperatures of the magma (at the time of intrusion) and the surroundings, allows prediction of the T–t history at any point. In practice, there are many assumptions involved in such predictions, but these can be tested and refined with reference to aureole assemblages. Contact metamorphic aureoles commonly show evidence for disequilibrium; even in inner zones, where high temperatures are attained and reaction rates are expected to be fast, the timescale for heating and cooling is shorter than that for regional metamorphism. For instance, the Christmas Mountains aureole (Joesten, 1983) affects limestones with chert nodules; these nodules are rimmed by contact metamorphic assemblages, but did not react completely. Other aureoles, such as Ballachulish in Scotland (Voll et al., 1991), appear to have pelites equilibrated at the peak temperature attained in each zone. Here, the initial fine grain size of the regionally metamorphosed chlorite slates may have aided fast reaction. In quartzites in the same aureole there is evidence for disequilibrium during partial melting (Holness & Clemens, 1999), highlighting the importance of water supply if input of H2O is required for reaction. In general, dry coarse-grained rocks are less likely to equilibrate and therefore carry more information on the kinetics of reactions during contact metamorphism. In this paper, we describe how coarse-grained amphibolite-facies pelites responded to contact metamorphism in the aureole of the Ross of Mull pluton, Scotland. We show how disequilibrium is prevalent in the low-grade zones, and is marked even at the highest grades.
REGIONAL SETTING OF THE ROSS OF MULL PLUTON

The Caledonides in Scotland include a large area of regionally metamorphosed rocks: the Moinian Supergroup, deposited in the Precambrian (Harris, 1995). It was regionally metamorphosed prior to closure of the Iapetus suture, at c. 500 Ma, although the timing of that event remains controversial. The Moinian Supergroup also appears to have a Precambrian metamorphic history (Harris, 1995). The protoliths were dominated by clastic rocks, mainly arkoses and sandstones with subordinate pelites and thin limestones, but also included sheet-like bodies of basic rocks. Most of these rocks are now at amphibolite facies, the more psammitic protoliths giving rise to biotite schists and gneisses, and the pelites containing more varied mineral assemblages. After the closure of the Iapetus Ocean, a suite of late-orogenic granites and intermediate plutonic rocks was intruded into these already deformed and metamorphosed sediments. The Ross of Mull pluton is one such, intruded at 414 ± 3 Ma (Halliday et al., 1979) and ranging from monzogranite to granodiorite in composition (British Geological Survey, 1999).

The island of Mull contains a small isolated area of Moinian metasediments (Bosworth, 1910; Holdsworth et al., 1987). These are bounded on the NE by downfaulted Tertiary flood basalts, on the west by the Ross of Mull pluton, and to the north and south by sea (Fig. 1). The Ross of Mull pluton is itself bounded to the west mainly by sea, but small islands just off the east shore of the island of Iona are granite (Potts et al., 1995). The nearby country rocks on Iona are psammites and pelites of the Iona group, which are not correlated with the Moinian Supergroup—faulting and shearing in the Sound of Iona are postulated to account for the (pre-intrusion) juxtaposition of different units (Potts et al., 1995). Hornfelsing of the Iona rocks (British Geological Survey, 1999) shows that they were affected by the Ross of Mull pluton. Within the Moinian inlier the dominant rock type is a biotite–quartz–feldspar schist or gneiss sometimes with garnet, interpreted as meta-arkose, psammitite or pelitic variants of these depending on abundances of feldspar and mica. Basic sheets (now garnet amphibolites), calc-silicate lenses and pelites (now gneisses with some or all of garnet, biotite, kyanite and muscovite) are subordinate. Upright folds in the inlier trend NE–SW. There are two main outcrops of pelitic lithology: the main one is a band that trends NE–SW across the middle of the inlier [within the Ardalanish Striped and Banded Formation of Holdsworth et al. (1987)]; the other occurs in the NW of the inlier adjacent to the pluton. The response of these metapelites to the contact metamorphism is the focus of this study. Because the main pelite band is oblique to the roughly north–south-trending pluton contact, it exhibits diverse contact metamorphic mineral assemblages that partly overprint the regional metamorphic assemblages. The thermal metamorphic assemblages were first described by Bosworth (1910), Cunningham-Craig (1911) and MacKenzie (1949), but there are no more recent published studies. The aureole was mentioned briefly by Kerrick (1990, p. 278) but it should be noted that the scale bar on his fig. 9.12 was mistakenly labelled as ‘10 km’ whereas it is in fact 1 km.

The intrusion has an east–west outcrop width of 7 km (British Geological Survey, 1999). Gravity modelling (Beckinsale & Obradovich, 1973) has been interpreted to show that it is an east-dipping slab with a true thickness of 3–4 km, but other cross-sections suggest a more vertical-sided body with true width of the order of 7 km (British Geological Survey, 1999). There is little evidence of plastic deformation of the country rocks during intrusion: instead, rafts of country rock quartzite, dismembered but unrotated, and mappable ‘ghost stratigraphy’ within the granite suggest that the pluton attained its final position by stopping. Although dominated by granitoids, the intrusion is not all granite. Mafic enclaves in the west provide evidence for magma mingling, and there are more dioritic parts to the intrusion. It is possible, then, that the thermal aureole was due, in part, not to the granite but to more mafic and therefore hotter magmas.

In subsequent sections we first outline the variety of regional metamorphic mineral assemblages found in the main pelite band at its NE end, where there is no visible evidence of contact effects. We then document the new mineral chemistries that give rise to different mineral assemblages. The pluton is approached, and diagnose the main reactions that are giving rise to these. Subsequently we discuss the effect of the protolith on reaction kinetics.

REGIONAL METAMORPHISM

In detail, the pelite band shows a variety of bulk-rock chemistries that give rise to different metamorphic assemblages (Mangan, 1996). Four regional metamorphic assemblages have been identified (Table 1). Assemblage 1 is the divariant Grt–Bt–Ky–Ms–Qtz assemblage in KFMASH [all abbreviations after Kretz (1983)] and assemblages 2 and 4 are related trivariant assemblages, each with one phase from the above list absent, reflecting variable bulk compositions. Assemblage 2 lacks muscovite (so is poor in K), assemblage 3 lacks garnet (low Fe/Mg) and assemblage 4 lacks kyanite (poor in Al). All assemblages contain plagioclase and rutile reflecting the non-KFMASH components Na, Ca and Ti. Garnet is typically 2–5 mm across, biotite 2 mm and bladed
kyanite porphyroblasts range up to 5 cm × 1 cm. Micas generally form a strong foliation. Significant amounts of tourmaline are found in places within the kyanite gneisses—there is no textural evidence that suggests that this is related to contact metamorphism. Euhedral tourmalines are in places included within kyanite grains and therefore the tourmaline is considered to have crystallized during regional metamorphism, possibly as a result of metasomatism. The mineral chemistry of the regional assemblages provides a template for understanding their modification as the aureole is approached. The complete dataset of analyses from 29 samples outside and inside the aureole may be downloaded from the *Journal of Petrology* website at [http://www.petrology.oupjournals.org/](http://www.petrology.oupjournals.org/).

Table 2 shows selected analyses but only from inside the aureole. Figure 2 plots mineral compositions and tie-lines between minerals for several samples from outside the aureole. The absence of crossing tie-lines shows that these regional assemblages are compatible at a single pressure and temperature.

The regionally metamorphosed rocks show evidence for disequilibrium: plagioclase varies slightly in composition within individual rocks, and garnet is occasionally partly replaced by retrogressive chlorite. Bearing this in mind, we have constrained the regional conditions using Fe–Mg exchange between garnet and biotite (Ferry & Spear, 1978), giving temperatures in the range 610–700°C for a nominal pressure of 8 kbar. There is

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**Fig. 1.** Geological setting of Ross of Mull pluton and approximate location of isograds, shown as black roughly north–south-trending lines. Thick grey lines indicate kyanite-bearing bands that terminate at inferred faults shown by black dashed lines. Isograds can be defined only within these bands, and then only approximately. Dots indicate sample localities: those labelled are referred to in this paper or the Electronic Appendix; others were discussed by Mangan (1996).
Table 1: Key features of regional and contact metamorphic assemblages

<table>
<thead>
<tr>
<th>Ass.</th>
<th>Regional Zone I</th>
<th>Zone II</th>
<th>Zone III</th>
<th>Zone IV</th>
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<tr>
<td>1</td>
<td>Grt Bt Ky Ms</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td></td>
<td>Fine-grained Ms replacing Ky; sporadic Chl; e.g. Ky2, Ky5, Ky16, Ky21, Ky25, Ky39, Ky46</td>
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<td></td>
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<tr>
<td></td>
<td>Ky rimmed by Crd + Ms; e.g. Ky23</td>
<td></td>
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<td></td>
<td>Grt rimmed by Crd + Pl + Bt; Ky rimmed by Crd + Ms; And patches in Ky; Rt rimmed by Ilm; e.g. 70411-10 (Fig. 3a), 70413-13 (Fig. 3f)</td>
<td></td>
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<td></td>
<td>Grt pseudomorphed by Crd + Pl + Bt + Ilm; And rimmed by Crd + Kfs + Bt; Ky rimmed by Crd + Ms; And patches in Ky; Rt rimmed by Ilm; e.g. 70414-10 (Fig. 3d), 70411-2 (Fig. 3c), Kb</td>
<td></td>
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<td></td>
<td>Inclusion-free Grt; relict Bt contains Sil needles; Sil prisms in matrix; And rimmed by Crd; Kfs in matrix; Ilm; e.g. 717B (Fig. 3h), C15 (Fig. 3i, j), 70414</td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>3</td>
<td>Bt Ms Ky</td>
<td></td>
<td></td>
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<tr>
<td></td>
<td>e.g. Ky49, Ky51</td>
<td></td>
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<td></td>
<td>Ky rimmed by Crd + Ms; e.g. B7 (Fig. 3g)</td>
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<td></td>
<td>Ky rimmed by Crd + Ms; And patches in Ky; e.g. Kc, Kf2, Ky61, Ky62, Ky54 (Fig. 3b), Kg (Fig. 3d)</td>
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<tr>
<td></td>
<td>Grt pseudomorphed by Crd + Pl + Bt + Ilm; relict Bt contains Sil needles; prismatic And in matrix; Kfs in matrix; Ms also present as large irregular masses; e.g. Be, Be, Bg</td>
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<td></td>
<td></td>
</tr>
</tbody>
</table>

Underlined are the key KFMASH minerals characterizing each regional assemblage. Empty fields indicate no information. All rocks contain quartz and plagioclase in addition to the tabulated phases, and may contain rutile. In bold are sample numbers for which chemical data are provided here or in the Electronic Appendix. In italic are sample numbers relating only to photomicrographs. A question mark before a sample number indicates that there is uncertainty about the original regional assemblage; in Zone IV samples the regional assemblages cannot be identified.
no ilmenite in the rocks that have not been thermally overprinted, but rutile is present. The mineral pair rutile + garnet can be used to give a minimum pressure estimate based on the GRAIL geobarometer (Bohlen et al., 1983). The most Fe-rich regional garnet found has Fe/(Fe + Mg) = 0.84 and this gives $P > 8$ kbar for a nominal $T = 650^\circ$C. The pressure-sensitive equilibrium between grossular, anorthite, plagioclase and quartz (GASP; Hodges & Crowley, 1985) could be applied in principle, but plagioclase chemistry is too variable to be used for an equilibrium calculation. We have found staurolite (rarely) in the aureole rocks, but also far from the intrusion, as documented by Holdsworth et al. (1987). It is unlikely to be a contact metamorphic mineral, and textures suggest that it was part of the regional assemblage, and cofacial with the garnet amphibolite sheets.

### CONTACT METAMORPHISM

Four metamorphic zones (I–IV) have been distinguished using the pelite outcrops; unfortunately, these cannot be mapped in the surrounding biotite schists (Table 1, Fig. 1). In the lower-grade zones, relict regional mineral assemblages can still be identified, but at the highest grade this is not possible. Textures characterizing the zones include:

- **Zone I**: cordierite–muscovite coronas round kyanite;
- **Zone II**: cordierite–muscovite coronas round kyanite; cordierite–biotite–plagioclase–ilmenite in and around garnet; partial replacement of kyanite by andalusite;
- **Zone III**: K-feldspar, andalusite and fibrolite;
- **Zone IV**: cordierite around garnet; euhedral garnet; K-feldspar; prismatic sillimanite.

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### Table 2: Representative mineral compositions for Zones I–IV

<table>
<thead>
<tr>
<th>Zone:</th>
<th>Cordierite (to 18 O)</th>
<th>Garnet (to 12 O)</th>
</tr>
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<tbody>
<tr>
<td>Sample:</td>
<td>Ky57</td>
<td>K32</td>
</tr>
<tr>
<td>Reg. assem.:</td>
<td>4</td>
<td>3</td>
</tr>
<tr>
<td>Textural setting:</td>
<td>Corona round Grt</td>
<td>Corona round Ky</td>
</tr>
</tbody>
</table>

| SiO$_2$ | 48.13 | 48.76 | 47.48 | 48.11 | 46.73 | 47.35 | 36.25 | 36.83 | 37.46 | 36.57 | 36.51 | 37.70 |
| TiO$_2$ | 0.02 | 0.04 |
| Al$_2$O$_3$ | 32.24 | 32.99 | 32.16 | 32.27 | 32.11 | 32.48 | 21.00 | 20.73 | 20.82 | 21.05 | 20.91 | 21.75 |
| FeO | 9.98 | 7.00 | 11.21 | 10.00 | 10.10 | 12.09 | 5.23 | 5.23 | 5.23 | 5.23 | 5.23 | 5.23 |
| MnO | 0.30 | 0.38 | 0.65 | 0.23 | 0.37 | 0.22 | 5.23 | 5.23 | 5.23 | 5.23 | 5.23 | 5.23 |
| MgO | 7.09 | 8.95 | 6.46 | 7.09 | 7.03 | 6.08 | 5.86 | 5.86 | 5.86 | 5.86 | 5.86 | 5.86 |
| CaO | 0.08 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 |
| Na$_2$O | 0.80 | 0.46 | 0.20 | 0.64 | 0.53 | 0.80 | 0.19 | 0.37 |
| K$_2$O | 0.02 | 0.02 |
| Total | 97.74 | 98.96 | 98.29 | 98.56 | 97.15 | 98.79 | 98.09 | 99.25 | 100.41 | 100.86 | 99.75 | 100.09 |

**Si**

| Ti | 4.11 | 4.98 | 4.98 | 5.01 | 4.94 | 4.95 | 2.98 | 2.98 | 3.01 | 2.95 | 2.97 | 2.97 |
| Ti | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 | 0.25 |
| Al | 4.01 | 3.97 | 3.97 | 4.01 | 4.00 | 4.01 | 2.03 | 1.98 | 1.97 | 2.00 | 2.00 | 2.03 |
| Fe | 0.86 | 0.60 | 0.87 | 0.89 | 1.06 | 1.06 | 2.07 | 2.18 | 2.31 | 2.34 | 2.34 | 2.34 |
| Mn | 0.03 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 | 0.04 |
| Mg | 1.07 | 1.01 | 1.07 | 1.11 | 0.95 | 0.95 | 0.32 | 0.36 | 0.36 | 0.36 | 0.36 | 0.36 |
| Ca | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 |
| Na | 0.09 | 0.13 | 0.11 | 0.11 | 0.11 | 0.11 | 0.11 | 0.11 | 0.11 | 0.11 | 0.11 | 0.11 |
| K | 5.94 | 5.94 | 5.94 | 5.94 | 5.94 | 5.94 | 5.94 | 5.94 | 5.94 | 5.94 | 5.94 | 5.94 |

**Mg no.**

| alm | 69.0 | 70.0 | 70.0 | 70.0 | 70.0 | 70.0 | 50.0 | 70.0 |
| spss | 12.0 | 6.0 | 6.0 | 6.0 | 6.0 | 6.0 | 6.0 | 6.0 |
| py | 10.7 | 13.4 | 11.9 | 12.7 | 12.7 | 12.7 | 3.5 | 9.0 |
| gfs | 8.3 | 9.5 | 9.5 | 9.5 | 9.5 | 9.5 | 9.5 | 9.5 |

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We now describe these and other textures and interpret them in terms of simplified stoichiometric reactions between mineral end-members. Our main assumption in doing this is that coronas represent reaction products, with the central mineral being a reactant. Ambiguity arises for complex coronas where ‘distant’ minerals must have been involved as reactants. This first stage of analysis allows us to approximate $P-T$ conditions in the aureole. We give several balanced reactions that summarize what is happening in the aureole. Some of these reactions appear as equilibria on $P-T$ grids; others are metastable, partly because the regional assemblages were so far from equilibrium when contact metamorphism occurred. We return in a subsequent section to a more detailed discussion of the reactions and textures.

**Zone I**

In rocks bearing regional kyanite, this mineral is rimmed by relatively fine-grained cordierite and muscovite aggregates. Because all such rocks contain biotite, the principal reaction involved is

$$\text{Ky} + \text{Bt} + \text{Qtz} + eW \rightarrow \text{Crd} + \text{Ms}. \quad (R1)$$

Here we use the symbol $eW$ to indicate that some water might be required, depending on the non-stoichiometric water content of the cordierite, but apart from this the reaction is water-conserving. We will use the symbol $W$ when phases containing different amounts of stoichiometric water are involved. Rarely, some garnet breakdown products are observed, but these are more...
characteristic of Zone II and are, therefore, discussed in the next section.

Zone II

As in Zone I, when kyanite is present, it shows cordierite–muscovite coronas or more pervasive replacement by those minerals (Fig. 3b). Regional biotite is always present and we infer the operation of (R1). Closers to the pluton within Zone II, it is evident from field and thin-section observations that these coronas are thicker; at the hillock of Dun Fuinn (Fig. 1) some kyanites are almost entirely pseudomorphed by the new minerals. This pattern of increasing corona width towards the pluton is discussed later. The cordierites in these coronas are commonly pinitized, but some robust analyses of cordierite were obtained (Fig. 4; Table 2). Although the coronas are dominated by muscovite and (pinitized) cordierite, these are intergrown with small amounts of decussate biotite inferred to be new. This has a different composition from the large aligned biotite flakes inherited from the regional metamorphism.

In addition, in the field, blue kyanite blades are seen to contain irregular masses of flesh-pink granular andalusite (Fig. 3c). This is evidence for the reaction

\[
\text{Ky} \rightarrow \text{And.} \quad (R2)
\]

As the pluton is approached within Zone II, it is evident from field observations that andalusite is more abundant: it occurs in a greater proportion of relict kyanites, and occupies, on average, more of each relict kyanite [Fig. 3d; see also fig. 112 of Yardley et al. (1990)]. This pattern accords with the prediction that higher peak temperatures will lead to a greater overall transformation, for any thermally activated reaction. In thin section, these masses of andalusite show undulose extinction (Fig. 3d). The kyanite cleavage is subdued or lost within these masses. The kyanites are commonly slightly kinked, but this is probably due to late regional deformation, as it is observed far from the pluton. Andalusite with strong undulose extinction is sometimes enclosed within undeformed kyanite. Although the undulose extinction gives the appearance of subgrains, we infer that it is not due to imposed deformation, because then the kyanite would also be strained. Instead, it is a consequence of the mechanism of andalusite growth.

Another texture characteristic of Zone II is rutile rimmed by ilmenite. Figure 4 shows that new biotites are more Fe-rich than old biotites so this mineral cannot be the source for the Fe in ilmenite. In addition, rutiles are not rimmed in Assemblage 3 rocks in which garnet is absent. This suggests the operation of

\[
\text{Alm} + \text{Rt} \rightarrow \text{Ilm} + \text{Qtz} + \text{Al}_2\text{SiO}_5'. \quad (R3)
\]

This resembles the GRAIL reaction used in geobarometry (Bohlen et al., 1983) but here we do not suggest it necessarily led to the production of more aluminium silicate as a phase, but instead that the Al and Si could have been involved in other reactions discussed below.

Zone II shows large regional garnets rimmed by aggregates of cordierite, relatively fine-grained biotite, plagioclase, ilmenite and some muscovite (Fig. 3a). These are present in all garnet-bearing lithologies, both in the presence and absence of regional kyanite. The plagioclases in these coronas are more calcic than those in the groundmass. Cordierites are not always pinitized and the relationship between their compositions and those of new and relict biotite is shown in Fig. 4. The corona biotites

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**Fig. 2.** (a) Thompson AFM diagram for regional assemblages. This has been drawn by projecting from pure muscovite; this is not completely valid, as the actual white mica is somewhat phengitic. Garnet contains some grossular, which is implicitly projected from, and biotite contains Ti, implying an implicit projection from rutile. The diagram does, however, illustrate the compatibility between the assemblages and forms a useful template for displaying disequilibrium assemblages in the aureole. Pale grey areas indicate trivariant assemblages; dark grey area indicates a quadrivariant assemblage. (b) Projection from kyanite (and quartz, water) onto KFM plane in KFMASH for Ky-bearing assemblages, including assemblage 2, which cannot be shown on the Thompson AFM diagram.
Fig. 3. Textures of aureole rocks (see Table 1 for setting in terms of original assemblage and aureole zone). Width of each photomicrograph is 3.95 mm. (a) Sample 70411-10, plane-polarized light (PPL). Garnet rimmed by new biotite, cordierite and ilmenite (note also biotite in cracks). (b) Sample Ky54, PPL. Pseudomorphed elongate kyanite, showing only two high-relief relics at either end. (c) Sample 70411-2, cross-polarized light (XPL). Andalusite ‘island’ within kyanite; note fingers of andalusite parallel to (001) of kyanite. (d) Sample Kg, XPL. Andalusite pseudomorph after kyanite. Andalusite shows complex extinction, and a corona of cordierite and muscovite. (e) Sample 70413-10, PPL. Garnet completely pseudomorphed by plagioclase and cordierite (colourless), biotite, and ilmenite (black). (f) Sample 70413-13, PPL. Prismatic andalusite cross-cutting regional biotite foliation, surrounded by and including flakes of regional biotite. (g) Sample B7, PPL. Fibrolite mats associated with and probably replacing biotite. (h) Sample 117B, PPL. Garnet associated with fibrolite (top right), prismatic sillimanite and other colourless phases as shown. (i) Sample Ct5, PPL. Small anhedral garnet partly rimmed by cordierite. (j) Sample Ct5, XPL. Same field of view as (i). (k) Field photograph of kyanite blades (pale blue) partly or completely pseudomorphed by andalusite (pale pink), Zone II, Dun Fuinn. Coin for scale, diameter 2 cm. Yellow, grey and white patches are lichen.
are consistently more Fe-rich, Al-rich and Ti-poor than regional biotites from the same thin section. They are also more Fe-rich than the new cordierites, which is to be expected if these two new phases are in equilibrium with each other. Both cordierite and plagioclase could be produced, in kyanite-bearing assemblages, by

\[
\text{Grt} + \text{Ky} + \text{Qtz} + \text{eW} \rightarrow \text{Crd} \quad \text{(R4)}
\]

\[
\text{Grs} + \text{Ky} + \text{Qtz} \rightarrow \text{An.} \quad \text{(R5)}
\]
This zone is characterized by the coexistence of aluminium silicate with K-feldspar. K-feldspar appears, together with cordierite, in coronas around andalusite masses (pseudomorphs after kyanite), and within relict regional biotites and muscovites (although it is not interpreted as relict itself). Andalusite is present not only as irregular masses with undulose extinction in pseudomorphs after kyanite, but also as prismatic grains in the matrix, commonly cutting across foliated (relict regional) biotite and including that phase (Fig. 3f). The prismatic matrix andalusite is, in places, slightly salmon pink and pleochroic in thin section, indicating some Fe\(^{3+}\) and/or Mn\(^{3+}\) (Deer et al., 1982). The andalusite in pseudomorphs has no detectable levels of those elements. The presence of K-feldspar as well as this matrix andalusite (which is not associated with kyanite) suggests

\[
\text{Ms} + \text{Qtz} \rightarrow \text{Kfs} + \text{And/Sil} + W. \quad (R9)
\]

Two other reactions that can produce K-feldspar, and that may have operated locally, are

\[
\text{Ms} + \text{Bt} + \text{Qtz} \rightarrow \text{Kfs} + \text{Crd} + W \quad (R10)
\]

\[
\text{Ky}/\text{And/Sil} + \text{Bt} + \text{Qtz} \rightarrow \text{Kfs} + \text{Crd} + W. \quad (R11)
\]

Prismatic matrix andalusite could also have been produced by

\[
\text{Crd} + \text{Ms} \rightarrow \text{And} + \text{Bt} + \text{Qtz} + \text{W}. \quad (R12)
\]

Fibrolite is present in Zone III as mats within relict regional biotite (Fig. 3g). It is again not straightforward to identify the reaction producing this texture. Reactions (R10) and (R11) both indicate biotite as a reactant but these do not produce Al\(_2\)SiO\(_5\). Fibrolite is, despite the large differences in Al between the two phases, found within biotite in many metamorphic terrains and this is probably due to ease of nucleation there (Chinner, 1961; Kerrick, 1990, p. 276). It has been proposed that fibrolite is produced from biotite by leaching of K, etc. (Kerrick, 1987), but we find no evidence for fluid flow here. We favour a model for fibrolite and matrix sillimanite based on reaction (R9), where biotite is involved as a preferred nucleation site (see Carmichael, 1969).

Aggregates of cordierite, plagioclase, biotite and ilmenite are interpreted as pseudomorphs after garnet (Fig. 3e). In this zone, then, regional garnet and kyanite have almost completely disappeared; aluminium silicate and K-feldspar are stable, but regional muscovite and biotite persist. Figure 5 shows coexisting cordierite and biotite compositions in Zone III.

**Zone IV**

The main boud of pelites does not extend all the way to the pluton in the SW of the Moinian inlier and is probably faulted out (Fig. 1). To the NW, however, outcrops of similar pelites are found adjacent to, and as large xenoliths or roof pendants within, the pluton. These are assigned to Zone IV. Many of the textures in Zone IV...
resemble those in Zone III but in addition, prismatic sillimanite occurs [see MacKenzie & Guilford (1980, p. 15)], as do small garnets. Prismatic sillimanite is found in two textural settings: in 1–2 cm andalusite-rich ‘knots’ clearly visible in outcrop, and in the matrix. The knots themselves carry rare kyanite patches. Our favoured explanation of the knots is that they are pseudo-morphs after kyanite. Although they are equant rather than having the elongate outline of regional kyanite, we can think of no other way to obtain such large patches of aluminium silicate, and the rarely preserved kyanite must surely be a relict regional phase. Sillimanite may be produced by the direct reaction

\[
\text{And} \rightarrow \text{Sil} \quad (\text{R13})
\]

as well as by reaction similar to that in Zone III [reaction (R9)]. The small garnets can be markedly euhedral; this is visible in the field in the outcrops c. 20 m east of Bendoran Cottage and recorded in thin section [e.g. Brearley & Champness (1986, fig. 1b)]. These do not show signs of reaction rims or growth of new minerals along cracks. The small garnets are interpreted as new contact metamorphic products, from one of the reactions

\[
\text{And/Sil} + \text{Bt} + \text{Qtz} \rightarrow \text{Grt} + \text{Crd} + \text{Kfs} + \text{W} \quad (\text{R14})
\]

\[
\text{And/Sil} + \text{Bt} + \text{Qtz} \rightarrow \text{Grt} + \text{Crd} + \text{Kfs} + \text{L} \quad (\text{R15})
\]

It should be noted that the reaction can be balanced using either water (W) or melt (L) on the right-hand side. The picture is complicated by the occurrence, also in outcrops east of Bendoran Cottage and in a roof pendant west of there, of anhedral garnets with various coronas including biotite (Fig. 3h) or commonly entirely cordierite. We deduce these are relict regional garnet. In Zone III regional garnets are commonly entirely pseudomorphed, and one might expect the same in Zone IV. However, given the complexities of kinetics, this prediction is not robust. We propose that these anhedral garnets are resorbed regional metamorphic garnets. The resorption may have remained incomplete because, for example, some coronas are single crystals of cordierite. Corona growth would then have involved lattice diffusion through cordierite, which is much slower than grain boundary diffusion in the polycrystalline coronas of Zone II. These issues are discussed further in a subsequent section.

The presence of two or three Al2SiO5 polymorphs indicates the extent of disequilibrium. Further evidence for this is found in the variable chemistry of phases such as cordierite. Figure 6 shows that cordierites adjacent to new garnets have distinct chemistries from those further away in the matrix, which are more Fe-rich.

<table>
<thead>
<tr>
<th>PRESSURE–TEMPERATURE CONDITIONS IN THE ROSS OF MULL PLUTON AUREOLE</th>
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<td>P–T grid to be used</td>
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The extent of disequilibrium means that it would not be easy to use our data to propose or modify P–T grids and equilibria related to low-pressure metapelite assemblages. Instead, we use the grid that has been refined over many years using various methods and data sets.
years by Pattison and co-workers (Pattison & Harte, 1997), which we will refer to as the PH97 grid (Fig. 7). We remark on specific equilibria now, and give reasons for this choice.

The stability limits of Aluminium silicate + Biotite + Qtz in KFMASH

Powell & Holland (1990) and Xu et al. (1994) calculated KFMASH P–T grids using an internally consistent thermodynamic database (Holland & Powell, 1990). These grids predict that, although biotite– sillimanite–quartz can be stable, the assemblage biotite–andalusite–quartz is unstable in KFMASH, being represented instead by cordierite–muscovite–staurolite or, at higher temperatures, cordierite–muscovite–garnet. However, biotite–andalusite–quartz occurrences are common in many metamorphic terrains and this is acknowledged as a problem with the thermodynamic database (Powell & Holland, 1990). Occurrences of muscovite–cordierite–staurolite–biotite, which, if in equilibrium, would indicate the stability of cordierite–muscovite–staurolite, are instead argued to be polymetamorphic (Pattison et al., 1999). Therefore data from natural systems are more compatible with the PH97 grid.

The equilibrium Crd + Ms = And + Bt + Qtz + εW (R12)

This is of great importance in understanding low-pressure assemblages in metapelites. There is no doubt that the right-hand side is favoured for more Fe-rich compositions, but alternative slopes for isopleths of this reaction have been proposed. Using data from many metamorphic aureoles, Pattison et al. (2002) proposed a negative slope, so that the stability field of aluminium silicate plus biotite increases with rising temperature. However, Powell & Holland (1990) and Xu et al. (1994) calculated a positive slope for the Mg end-member reaction using an internally consistent thermodynamic database (Holland & Powell, 1990). A more recent version of that database, and other databases, yields different positions but similar positive slopes (Pattison et al., 2002). Thermodynamic data have been revised in accordance with field constraints (Pattison et al., 2002) and a new KFMASH grid has been calculated from these data. The predicted grid for subsolidus phase relationships in the absence of graphite, and using the Pattison (1992) triple point, was given by Pattison et al. (2002, fig. 10a). This is very similar to the PH97 grid, except that the equilibrium (R12) in their diagram is contoured in terms

Fig. 7. P–T grid (Pattison & Harte, 1997). Black lines and labels indicate univariant KFMASH equilibria; black dashed lines and italic labels, isopleths for divariant KFMASH reactions; Mgxy, cordierite Mg number for some isopleths; grey dotted lines, alternative positions for the And = Sil equilibrium (Richardson et al., 1969; Holdaway, 1971). Isopleths for (R16) are sketched in for completeness. Thick grey lines with arrows indicate approximate ranges of peak temperature for zones I–IV in the Ross of Mull aureole, assuming $P = 3.2 \text{kbar}$.
of biotite Mg-number rather than cordierite Mg-number. Using a cordierite–biotite $K_{D}$ of 0.57 (Pattison et al., 2002) shows that the isopleths in the two versions of the grid are not greatly dissimilar. The PH97 grid has the advantage that the isopleths are extended to different equilibria at higher temperatures. In summary, with regard to both this equilibrium and that discussed in the previous section, data from natural systems are more compatible with the PH97 grid.

Using the PH97 grid, then, the divariant assemblage Bt–And–Qtz–Crd–Ms is expected to show a cordierite composition varying (becoming more Fe-rich) with temperature (at fixed pressure) but not with bulk-rock chemistry; trivariant assemblages such as Bt–Qtz–Crd–Ms are expected to show cordierite compositions varying with bulk-rock chemistry but not temperature.

**The equilibrium Bt + And/Sil + Qtz = Crd + Kfs + W (R11)***

The PH97 grid predicts that, just up $T$ of the muscovite dehydration breakdown line (R9), for a given $P$ and $T$ there is a specific cordierite composition buffered by $\text{Al}_2\text{SiO}_5$, K-feldspar, biotite, quartz and water. This buffered cordierite composition becomes more Fe-rich as $T$ rises at fixed $P$ in accordance with the sliding reaction (R11):

$$\text{Bt} + \text{And}/\text{Sil} + \text{Qtz} \rightarrow \text{Crd} + \text{Kfs} + \text{W}.$$  

For a particular $P$ and $T$, cordierites more Mg-rich than this buffered composition may be in equilibrium with only two of sillimanite, biotite and K-feldspar. In unbuffered (trivariant) assemblages, for a given bulk chemistry, the cordierite composition would not vary at all as $T$ varies, if we acknowledge the negligible $T$ dependence of Fe–Mg exchange between biotite and cordierite (Pattison et al., 2002). Thus: the divariant assemblage Bt–And–Qtz–Crd–Kfs is expected to show a cordierite composition varying (becoming more Fe-rich) with temperature (at fixed pressure) but not with bulk-rock chemistry; trivariant assemblages such as Bt–Qtz–Crd–Kfs are expected to show cordierite compositions varying with bulk-rock chemistry but not temperature.

**The equilibrium Bt + Sil + Qtz = Grt + Crd + Kfs + W and related dehydration melting (R14) and (R15)***

This KFMASH univariant equilibrium appears on many grids (Xu et al., 1994; Pattison & Harte, 1997; Spear et al., 1999). At high pressures and temperatures it is metastable with respect to the dehydration melting reaction (R15) in which melt rather than water is produced (e.g. Holland et al., 1996; Cenki et al., 2002). In some studies Kfs appears on the left (low-$T$) side of that reaction (Carrington & Harley, 1995), although this does not affect the interpretations we give here. White et al. (2001) presented a grid for partial melting equilibria in NCKFMASH. The addition of Na and Ca makes the equilibrium (R15) divariant:

$$\text{Bt} + \text{Sil} + \text{Qtz} \pm \text{Pl} = \text{Grt} + \text{Crd} + \text{Kfs} + \text{L}.$$  

However, this and other divariant fields that correspond to KFMASH univariant melting reactions are narrow and close in position to the KFMASH univariant lines. This is illustrated in, for example, fig. 7 of White et al. (2001), where the divariant field of Bt–Sil–Qtz–Grt–Crd–Kfs–Pl–L is shown spanning just a few degrees at c. 740°C (at 5 kbar). Although this grid is very sophisticated, it is based on a thermodynamic dataset that predicts that equilibrium (R12) will favour muscovite + cordierite as $T$ rises (see, for example, the subsolidus part of White et al.’s fig. 7), which contradicts field data as argued in the previous section. The univariant line (R15) on the PH97 grid will be taken as an adequate approximation to the divariant region that would be present in NCKFMASH. We have sketched in isopleths for where the divariant reaction

$$\text{Bt} + \text{Sil} + \text{Qtz} = \text{Grt} + \text{Kfs} + \text{L}$$  

would appear on the PH97 grid—the slopes are constrained topologically [see Cenki et al. (2002), although those workers showed Kfs on the low-$T$ side of this equilibrium].

**Assumptions involved**

We will assume that the peak temperature at each point was attained at roughly the same pressure. This will not be true if the pluton and its aureole were being exhumed quickly, or if it has been substantially tilted, but we have no evidence for either of these scenarios. Although in some models the pluton is portrayed as a dipping sheet (as mentioned above), there is no suggestion it has been tilted. The errors involved here are likely to be smaller than those arising from other uncertainties. Our problem is to distinguish what parts of each texture are likely to represent equilibrium assemblages for each zone and to use a petrogenetic grid to constrain $P$ and $T$ using these assemblages, bearing in mind that grids are themselves subject to uncertainty and must be assessed pragmatically.

The previous section indicates various types of disequilibrium in every zone:

1. regional relict minerals may persist, probably in disequilibrium, but are relatively easy to recognize;
2. new minerals are likely to be in equilibrium with each other in a given corona but in the lower-grade
zones, with separate coronas around different regional porphyroblasts, equilibrium between coronas is not guaranteed;

(3) not only regional metamorphic minerals, but also minerals formed on the prograde contact metamorphic path may persist; for example, in Zone IV, prismatic sillimanite must be the peak $\mathrm{Al}_2\mathrm{Si}_3\mathrm{O}_9$ polymorph, yet andalusite, itself thermal, remains.

The presence of thermal andalusite implies pressure below the $\mathrm{Al}_2\mathrm{Si}_3\mathrm{O}_9$ triple point, itself placed at 3.8 kbar (Holdaway, 1971), 5-2 kbar (Richardson et al., 1969) or somewhere between these values on the PH97 grid [point (a) in Fig. 7]. As discussed above, andalusite is found in Zone III not only in kyanite pseudomorphs but also as euhedral grains in the matrix. The latter are interpreted as being a product of the breakdown reaction (R9). Sillimanite is, however, also present in Zone III and interpreted as due to the equivalent to (R9). We infer that the muscovite breakdown reaction began by producing andalusite (R9) and then, as temperature rose, produced sillimanite instead. This sequence suggests that pressure was below that of the intersection of (R9) with the $\mathrm{And} = \mathrm{Sil}$ line [point (b) in Fig. 7], placing us within Facies Series 1 of Pattison & Tracy (1991); however, we argue below that pressure was in fact a little higher, placing the aureole just within Facies Series 2.

**Zone II: extent of disequilibrium**

The most obvious evidence for disequilibrium is the coexistence of andalusite with relict kyanite; the transformation is incomplete for kinetic reasons. Additional evidence for disequilibrium comes from considering the minerals Crd, Ms, And, Bt and Qtz. In all rocks from Zone II that contain relict kyanite, new cordierite, muscovite and andalusite are present, together with relict biotite and varying amounts of new biotite. This suggests that although kyanite (and, where present, regional garnet) breakdown is not complete, the new stable assemblage would have been $\text{Crd--Ms--And--Bt--Qtz}$. Zone II is rather wide and within it the peak temperature must have varied. If this divariant assemblage is in equilibrium throughout Zone II then, as discussed in the ‘cordierite equilibria’ section above, cordierite composition should vary systematically with position (specifically, more Mg-rich compositions are predicted for higher-$T$ assemblages closer to the pluton). However, the variation of cordierite compositions with position in Zone II (Fig. 8) is not systematic. Moreover, the isopleths in Fig. 7 show that high pressures are required to stabilize Mg-rich cordierite ($X_Mg = 0.7$, for example) in equilibrium with aluminium silicate, biotite, quartz and either muscovite or K-feldspar. Such pressures would mean that andalusite would never be a stable polymorph in the aureole and we reach a contradiction. Finally, the more Mg-rich cordierites from Zone II in Fig. 8 are from coronas round relict kyanite (with variable amounts of transformation to andalusite) whereas the more Fe-rich cordierites are from coronas round garnet.

We propose that the range in cordierite and new biotite compositions in Zone II is purely a function of bulk-rock (or local textural domain) composition, not of metamorphic grade. We suggest that, given more time, each
of the analysed rocks from Zone II would have equili-
brated to one of the two trivariant assemblages 
Crd−Ms−Bt−Qtz or Crd−Ms−And−Qtz. Which one would have been attained depends on whether the bulk-
rock chemistry lies above or below the cordierite line in
Fig. 4. For rocks whose bulk chemistry lies below the 
cordierite line, the andalusite forming from kyanite is 
metastable—it would eventually have reacted with biotite 
and been completely consumed. We next argue that the 
thicknesses of coronas in Zone II show that aluminium 
silicate would have been completely consumed at equilib-
rium, for the analysed Zone II rocks. We may imagine a 
rock with a particular M/FM ratio of biotite in contact 
with regional kyanite, and quartz. Let us suppose that this 
rock is taken to a variety of peak temperatures but at 
much lower pressures than those of the regional event, 
and the two grains react to reach equilibrium. At suffi-
ciently low temperatures (the precise conditions being a 
function of the M/FM ratio), the equilibrium assem-
blage would be Crd−Ms−Bt: the regional kyanite 
would be completely replaced. At intermediate tempera-
tures, And−Crd−Ms−Bt would be stable. Some kyanite 
would react, but the rest would transform to andalusite. 
At high temperatures, And−Bt (or Sil−Bt) would be 
stable. The kyanite would transform to andalusite but 
no other reaction need occur. In other words, we would 
expect less Crd-producing reaction occurring at higher 
grades, and narrower coronas. In fact, there is a general 
trend towards wider coronas and, in the higher-grade 
parts of Zone II, regional kyanites are commonly com-
pletely pseudomorphed by cordierite from kyanite.
This trend in corona widths is then an effect of 
thermally activated reaction kinetics. Even though the 
driving force (free energy reduction) for reaction is less at 
higher grades, diffusion must be involved and is so sensi-
tive to temperature that this kinetic effect dominates.

Zone III: possible equilibrium
In Zone III the rocks contain, amongst other phases, 
sillimanite, K-feldspar, cordierite, new biotite and quartz.
We propose that the systematic variation with position of 
cordierite and biotite chemistry (Figs 5 and 6) supports the 
hypothesis that these five phases were in equilibrium, 
rather than the alternative that the cordierite chemistry is 
controlled by bulk-rock chemistry and at least one of the 
other phases listed is in disequilibrium. A previous section 
discussing (R11) emphasized that a systematic variation of 
cordierite towards more Fe-rich compositions is to be 
expected with rising temperature.

The cordierite composition at the lowest temperature 
within Zone III \( (X_{\text{Mg}} = 0.55) \) would then indicate the 
intersection of that cordierite isopleth with the muscovite 
breakdown line. This gives point (c) in Fig. 7, at 3.2 kbar.
At this pressure, muscovite would break down just outside 
the andalusite stability field (using the And = Sil equilib-
rium line of Fig. 7). This is an apparent contradiction to 
the argument that matrix andalusite in Zone III was 
produced by muscovite breakdown, and therefore at 
pressures below that of point (b). There are several ways 
to explain this. First, the position of the And = Sil line still 
remains debatable, and experimental studies give widely 
varying predictions (Richardson et al., 1969; Holdaway, 
1971). Second, in Zone III, sillimanite is present only as 
fibrolite. This can only be stable at higher temperatures 
than coarse-grained, defect-free sillimanite (Salje, 1986; 
Penn et al., 1999), and andalusite could have crystallized 
metastably if defect-free sillimanite could not nucleate.
Third, the matrix andalusite is sometimes coloured, indi-
cating presence of Fe\(^{3+}\) and/or Mn\(^{3+}\) that will enlarge its 
stability field at the expense of that of sillimanite. If 
nucleation of coarse-grained sillimanite were inhibited, 
the line Ms + Qtz = And + Kfs + W would extend 
metastably to higher pressures, but close to the stable 
equilibrium line Ms + Qtz = Sil + Kfs + W. The \( X_{\text{Mg}} = 0.55 \) isopleth would intersect this line at approx-
imately 3.2 kbar and 650°C.

Zone IV: significance of garnet
Zone IV contains anhedral garnets with cordierite co-
ronas. The chemistry of these garnets is broadly compara-
ble with that recorded from relict garnets in other zones,
and they are interpreted as relics. Euhedral garnets are 
interpreted as new because of their morphology. On the 
PH97 grid, garnet can appear up-grade in two ways.
One is by the divariant reaction (R16), but this can only 
account for the appearance of garnet in Fe-rich cordierite-
absent assemblages, so cannot apply here (see, e.g. 
Pattison & Harte, 1997, fig. 8). Alternatively, garnet 
appears by the univariant dehydration melting reaction 
(R15), predicted to be at 775°C if \( P \) was 3.2 kbar.

There are three points to discuss in relation to the 
proposed reaction. First, euhedral garnets have Mn-rich 
cores that would stabilize garnet at lower temperatures 
than those shown on the PH97 grid (which includes only 
KFMASH equilibria). Second, there is limited field evidence 
for large-scale partial melting in Zone IV. Third, the 
grid indicates that, near the dehydration melting reac-
tion, cordierite should have evolved towards much more 
Mg-poor compositions than are actually seen. We 
address these points below.

Effect of Mn on garnet stability
Given the disequilibrium present at all grades, the fact 
that Na and Ca will also have an effect, and uncertainties 
in thermodynamic data, we cannot be definite about the 
effect of Mn in garnet on (R14) and (R15). In 
MnKFMASH, there will be a divariant field of Bt + 
Sil + Qtz + Grt + Crd + Kfs + L, stabilizing garnet at
lower temperatures than for the univariant line. At 3.2 kbar the univariant dehydration reaction (R14) is metastable with respect to the dehydration melting line (R15). However, in MnKFMASH the assemblage Bt + Sil + Qtz + Grt + Crd + Kfs + W will have a divariant field stretching down temperature from the position of metastable (R14). To gain some insight regarding the effect of Mn we calculated the amount required to stabilize garnet in the assemblage Bt + Sil + Qtz + Grt + Crd + Kfs + W, which is divariant in MnKFMASH. We assumed Mn was present only in garnet, and used the thermodynamic database of Holland & Powell (1998). Because, as explained above, this dataset does not reproduce key features of the PH97 grid, we use this model only to estimate how much a given Mn content in garnet will shift the equilibrium (R14) down temperature, not to predict its actual position. The modelling was performed using the Vertex software package (e.g. Connolly & Petrini, 2002). Figure 9a shows how garnet composition is predicted to vary. A spessartine content of 30% could stabilize garnet 30°C below the univariant (R14). Figure 9b is a sketch of how Mn would vary for garnet in equilibrium with melt, based on the premise that the assemblage in equilibrium with W will be stable at lower temperatures than that in equilibrium with L, and that KFMASH univariant (R15) is at lower T than (R14). If garnet were in equilibrium with L not W then a 30% spessartine content would stabilize it only less than 30°C below the univariant line (R15). The PH97 grid shows that W cannot be in equilibrium with plagioclase + K-feldspar + quartz above c. 670°C. We do not think it is possible that all of zones III and IV were squeezed into the small temperature interval 650–670°C, and therefore use the PH97 grid to predict that melt was likely to have been present in Zone IV. Thus new garnet cores grew at a minimum temperature of 745°C and rims, with lower Mn, are likely to have formed less than 20°C below (R15), in other words at a minimum of 755°C.

In summary, Mn in garnet could have allowed growth to begin perhaps 30°C below the temperature of the univariant (R15), but T must have risen to not far below that KFMASH univariant as Mn-poor rims developed.

Evidence for melting

The previous section argues that melting is likely to have occurred during garnet formation in Zone IV. In the field, near the pluton, various textures involving granitoids and country rock can be described as migmatitic. Some of these involve decimetre- to metre-scale sharp-edged granitoid sheets, which are clearly intrusive (e.g. on the south coast at 367179, and in outcrops east of Bendoran cottage). Elsewhere (e.g. between Bendoran cottage and the main pluton), smaller leucosome patches with less distinct boundaries are present. These could be due to anatexis, but are not markedly developed in the outcrops that exhibit the most euhedral garnets. We propose that melt produced by anatexis permeated some distance, into nearby country rocks or into bodies of intruded magma. In the exposures east of Bendoran Cottage, there are several metre-scale fingers of granite present, so migration over large distances is not required. If melt did not move at all, it would have back-reacted with other phases on cooling. This would have involved consumption of garnet and destruction of the euhedral outlines. We cannot envisage how the prograde, euhedral garnets could have survived if melt had remained present everywhere it had been generated.

Cordierite compositions in Zone IV

Extrapolation of cordierite isopleths on the PH97 grid shows that cordierites should evolve to markedly
Mg-poor compositions before reaction (R15) is encountered. Such compositions have not been measured, but even in a single thin section cordierite shows a variety of compositions (Table 2) and is clearly not in equilibrium. We suggest that cordierite compositions realigned to an extent on the retrograde path in Zone IV.

DISCUSSION

Unusually marked disequilibrium and the role of water

We have outlined various arguments that demonstrate the extent of disequilibrium in this aureole: they are based on a direct interpretation of textures and on relating assemblages and chemistry to a published petrogenetic grid. At least two unusual factors contribute to the extent of disequilibrium, both aspects of the high grade of the protolith.

1. Coarse grain size means that diffusion distances are longer than in a fine-grained protolith.

2. The protolith, being high grade, did not carry many hydrous minerals. Some prograde reactions, however, required input of H2O. We have not measured the water content of cordierite, but assume there must be some. In many aureoles, previously low-grade protoliths undergo dehydration during heating. In contrast, assuming the K/H ratio in regional biotite and new muscovite is the same, contact metamorphic reactions such as (R1) in the Ross of Mull aureole would require addition of H2O. This might be rate limiting. There are three possible sources of H2O in addition to that in biotite. First, there are small amounts of chlorite in the regional assemblages. This relates to retrogression after the peak of regional metamorphism, and the breakdown of this chlorite could plausibly provide the relatively small amounts of H2O required for reactions such as (R1). Second, H2O will have been released in the higher-grade parts of the aureole (e.g. from breakdown of muscovite in Zone III). Third, water could have come from the crystallizing pluton. However, in Zone IV water activity is predicted to be less than unity, if the garnet-producing reaction is correctly identified. This is evidence against water from the pluton having permeated through the wall rocks visible at present. In the aureole as a whole, there is not much macroscopic or microscopic evidence for large-scale fluid movement. Because water as a phase is generally considered to speed up reaction rates, even if it is not required to balance the reaction, its absence here is perhaps another reason for slow reaction rates.

Relative diffusion rates

Textures give some insight into the relative diffusion rates for different elements. This assessment, although qualitative, is based on the premise that nucleation, and attachment or detachment of atoms at interfaces, is not rate-controlling. The Ky → And transformation is an exception: as the two minerals are adjacent, no diffusion is involved; the transformation must have been controlled by interface kinetics. The reaction (R1) involves both kyanite and biotite as reactants. Coronas are observed around kyanite, not around biotite; biotites not adjacent to kyanites are unaffected. This suggests that diffusion of K, Mg and Fe (supplied from biotite) was easier than diffusion of Al from kyanite. Other metamorphic studies have also inferred that Al was a slow-diffusing species. Rutile in the matrix, distant from ferromagnesian phases, is rimmed by ilmenite, again suggesting that Fe could diffuse over significant distances, with Ti relatively immobile.

The slow rate of Al diffusion would inhibit garnet breakdown by (R4)–(R6), all of which involve transport of Al from other phases. Instead, the reaction scheme for garnet breakdown in Zone II was dominated by reaction with rutile. This can be thought of as (R3) releasing ‘Al2SiO5’ from garnet with (R4) and (R5) locally consuming this within the garnet pseudomorph, or, effectively,

Ca,Fe,Mg Grt + Rt + eW → Crd + Pl + Ilm.

Rutile inclusions in garnet are commonly rimmed by or completely replaced by ilmenite, in contrast to those in kyanite that are well preserved. This corroborates the suggested reaction scheme.

Intrusion temperature and heat transfer

At what temperature were the country rocks when the Ross of Mull pluton intruded? K–Ar ages from the intrusion of 423 ± 4 Ma (biotite) and 416 ± 4 Ma (hornblende) might in principle be used to constrain its cooling history (Beckinsale & Obradovich, 1973); however, the biotite ages, being older than those of hornblende, are probably affected by excess argon. The hornblende age overlaps with the Rb–Sr intrusion age of 414 ± 3 Ma (Halliday et al., 1979), indicating only that the intrusion cooled through 500°C soon after emplacement. An aureole pressure of 3-2 kbar gives a depth estimate of c. 12 km (for a nominal density of 2800 kg/m³). A high regional geothermal gradient of 30°C/km would imply an ambient temperature of 350°C, which we regard as an upper limit. Simple thermal models of intrusion show that the peak aureole temperature, adjacent to the intrusion, will be comparable with or less than the average of intrusion and wall rock temperatures (Furlong et al., 1991). A Zone IV temperature of 750°C would require a magma temperature of 1150°C, which is far too hot for a granitic magma. Although the intrusion is composite, with more mafic components implying the involvement of hotter magmas, these are only exposed in the SW (British Geological Survey, 1999) and there is no evidence for them near the studied aureole rocks. Instead, magma may have passed through the currently exposed level for a
prolonged period, so that the wall rocks became hotter than a purely conductive model would predict. Latent heat of crystallization will also have served to raise wall rock temperatures substantially. Fluids expelled from the magma, however, are not thought to have passed through the exposed aureole rocks, and cannot therefore have contributed to heat transfer. It is likely that fluids were lost at structurally higher levels in the pluton system, which have now been removed by erosion.

CONCLUSIONS
The Ross of Mull aureole affected initially coarse-grained, dry rocks that had previously been regionally metamorphosed. Four metamorphic zones have been defined, but none show equilibrium textures or assemblages.

Zone I shows regional kyanite rimmed by cordierite and muscovite formed from reaction with biotite. Zone II shows the same texture with a greater extent of replacement; in addition, kyanite is partly replaced by andalusite. The extent of replacement increases towards the pluton. Garnet is partly replaced by cordierite, plagioclase and new biotite. Incomplete reaction is due to slow diffusion through coarse-grained material, and limited supply of new biotite. Incomplete reaction is due to slow diffusion through coarse-grained material, and limited supply of $H_2O$. The andalusite, although new, is itself metastable. We show that, for a range of bulk compositions, the equilibrium AFM assemblage would have been cordierite and biotite, without andalusite. Diffusion was too slow to allow complete reaction of kyanite/andalusite to cordierite and muscovite.

Zone III shows the occurrence of K-feldspar and fibrolite, as a result of the breakdown of muscovite. Garnets may be completely replaced by cordierite, plagioclase and new biotite, and kyanite is sporadically preserved. Zone IV, which occurs adjacent to the pluton and in rafts within it, shows prismatic sillimanite as well as fibrolite. In some locations anhedral garnets surrounded by cordierite are probably regional relics. Others nearby are euhedral with Mn-rich cores. We suggest that the euhedral garnets are formed by the reaction of biotite with sillimanite to produce cordierite, K-feldspar, garnet and melt. The Mn content is not sufficient to greatly reduce the temperature of this reaction. Preservation of euhedral garnet forms suggests that the melt moved away to some extent, otherwise it would have back-reacted with garnet.

This aureole is unusual for showing all three aluminium silicate polymorphs, and for other markedly disequilibrium assemblage and textures. This character is due at least in part to the coarse-grained and dry nature of the protolith. In contrast, many other Caledonian intrusions such as Ballachulish (Pattison & Harte, 1997) and Fanad (Edmunds & Atherton, 1971) affect low-grade protoliths, including chlorite slates and schists. Such rocks would not require input of water to prograde and, being fine grained, would react relatively fast. The extent of disequilibrium in the Ross of Mull aureole makes it unusually difficult to determine metamorphic conditions, but especially interesting for the study of reaction kinetics.

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SUPPLEMENTARY DATA
Supplementary data for this paper are available on Journal of Petrology online.

REFERENCES


