# Garnet zoning and re-equilibration in the Strontian area, Scotland

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ABSTRACT. In Moinian metasediments regionally metamorphosed to sillimanite grade, garnet has subsequently been re-equilibrated in the inner (cordierite-potash feldspar) zone of the aureole of the Strontian Granodiorite. Zoning profiles of garnet from the aureole, and from surrounding regional assemblages, show retrograde Mn-rich rims. Some regional garnets are also internally zoned. In the aureole as a whole, the partition measure for Fe and Mg between garnet and biotite shows a correlation with that for Ca between garnet and plagioclase. If interpreted in terms of a simple equilibrium model, this might suggest that pressure decreased with increasing temperature towards the granodiorite, but the trend can more plausibly be attributed to deviation from ideal solid-solution behaviour of Ca in garnet and plagioclase, with incomplete homogenization of garnet as a complicating factor in the outer parts of the aureole. From the difference in the temperature thresholds for garnet core equilibration, the timescale $t_2$ of the thermal event relative to the regional $t_1$ is estimated as $t_2/t_1 \simeq 10^{-1.1 \pm 0.7}$, consistent with emplacement of the intrusion at an early stage of regional cooling.

THE use in geothermometry and geobarometry of distribution coefficients between garnet and other minerals depends on some part of the garnet being assumed to have reached equilibrium with its surroundings. The interior of a growing garnet may become isolated from its environment as growth proceeds ('refractory' behaviour; Hollister, 1969), so that the rim of the garnet may give the best indication of peak metamorphic conditions, as found in staurolite- and kyanite-bearing assemblages by Ghent et al. (1979); at higher grades of regional metamorphism, above about 600 °C, homogenization seems to be the rule (Tracy et al., 1976; Woodsworth, 1977; Yardley, 1977), except where metamorphism is short-lived, for example during rapid uplift (Selverstone and Hollister, 1980). Homogenized garnets often develop retrograde zoning at their edges as they cool, so the core compositions should be used in estimating, for example, peak metamorphic temperatures from the partition of Mg and Fe with cordierite (Tracy et al., 1976). Formation of homogeneous garnets,

followed by retrograde zoning, also appears to be common in the inner, migmatitic parts of deep-seated aureoles (Ashworth and Chinner, 1978). These are often surrounded by garnet-bearing regional metamorphic terrains, and it is pertinent to examine the effects of the thermal event on garnets at a distance from the central intrusion. In the Strontian area, pelites which regionally carry garnet have been modified, with the production of cordierite, near the Strontian Granodiorite (Ashworth and Chinner, 1978). Garnet remained a stable phase, but its composition changed. Evidence is here presented for re-equilibration of Fe and Mg between garnet and biotite in the inner, cordierite-potash feldspar zone of the aureole.

The Strontian Granodiorite intrudes Moinian metasediments, mainly pelites and semipelites of the Glenfinnan Division, which probably underwent metamorphism in the Grenville Orogeny about 1.0 Ga ago (Brook et al., 1976) but have suffered further, Caledonian regional metamorphism (Brook et al., 1977, Table 3). The area studied (fig. 1) lies in the sillimanite zone of a kyanite-sillimanite regional sequence (Winchester, 1974). Further north and west, at lower metamorphic grade in the Morar Division, garnets of pre-Caledonian age show striking growth-zoning (Olimpio and Anderson, 1978). They become progressively homogenized in the kyanite and sillimanite zones further east (Anderson and Olimpio, 1977) where the grade of Caledonian metamorphism is higher (Powell, 1974). The regional sillimanite zone around Strontian presumably reflects Caledonian metamorphism, since the granodiorite, whose zircon U-Pb age is $435 \pm 10$ Ma (Pidgeon and Aftalion, 1978), was intruded while the rocks were still hot (Watson, 1964), so that its aureole consists of modified schists and gneisses rather than hornfelses.

*Observations.* Garnet is a frequent but minor constituent of the metasediments, occurring as scattered grains $\sim 1$ mm in diameter. The assemblages are listed in Table I. Potash feldspar occurs with sillimanite in rocks within a few km of the
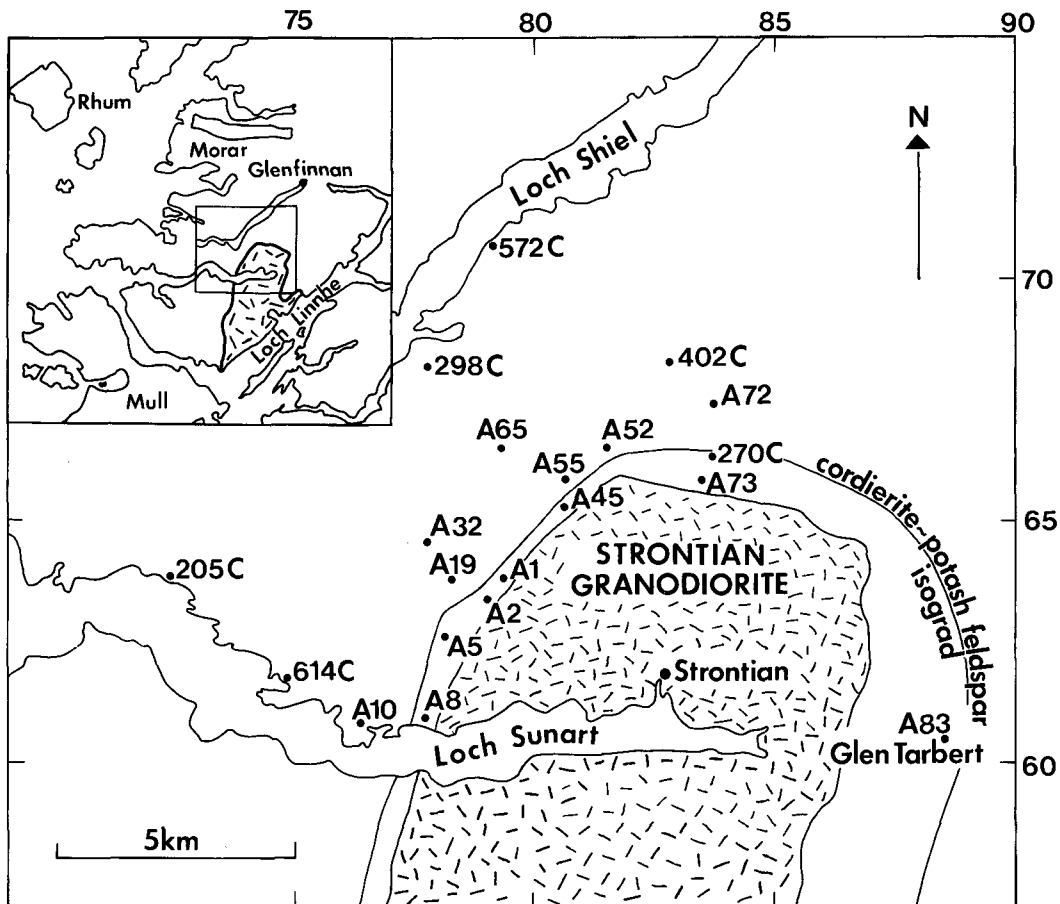
FIG. 1. Locations of samples. Inset shows the location of the area within the western Highlands of Scotland.

intrusion, and an inner cordierite–potash feldspar zone is defined by the incoming of cordierite (fig. 1). Migmatization by the thermal event is scarce, being suspected in only three of the samples from the cordierite–potash feldspar zone, and two from just outside that zone.

All but one of the fifteen garnets studied in detail have an outer rim in which $Mg/(Fe + Mg)$ decreases and Mn strongly increases, as exemplified in fig. 2. The exception is 205C (Table II), from a locality distant from the granodiorite.

The garnet from the most northerly locality, 572C (fig. 2a) has distinct internal zoning. An inner, relatively Ca-poor core is flanked by a zone of

TABLE I. *Assemblages*

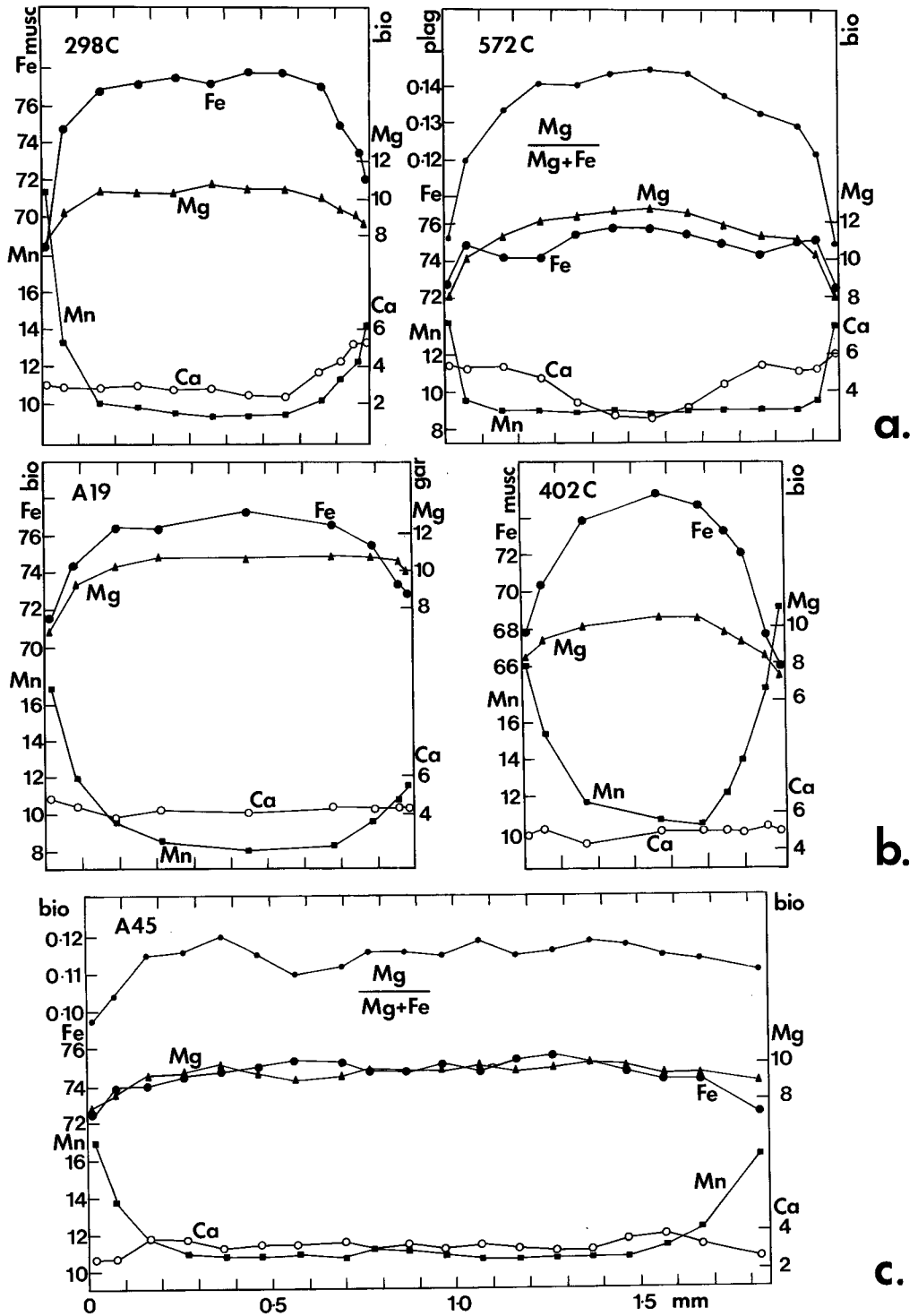| quartz | plag | bio | gar | sill | musc | K-feld | cord | andalusite | |
|--------|------|-----|-----|------|------|--------|------|-----------|---|
| x | x | x | x | x | (x) | x | x | | A1, A2, A5, A45, A83, 270C |
| x | x | x | x | x | (x) | x | x | x | A8 |
| x | x | x | x | x | | | | | A73 |
| x | x | x | x | x | (x) | x | | | A10, A19, A52, A55, A72 |
| x | x | x | x | x | x | x | | | 402C |
| x | x | x | x | x | x | | | | A32, A65, 205C, 298C, 572C, 614C |

(x) Regarded as secondary.

FIG. 2. Representative garnet zoning profiles. The mineral in contact with garnet at each end of a profile is indicated. Mg, Fe, Ca, and Mn are plotted in atomic percentages ($Mg + Fe + Ca + Mn = 100\%$). $Mg/(Mg + Fe)$ is plotted in atomic proportions. (*a*) Garnets from the regional sillimanite zone, distant from the intrusion. (*b*) Typical garnets from just below the cordierite–potash feldspar isograd. (*c*) Typical garnet from within the cordierite–potash feldspar zone.

TABLE II. *Representative electron-microprobe analyses of garnet and biotite*

| | Garnet | | | | | | | Biotite | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | A45 | | A19 | | 205C | | | | | |
| | Core | Rim | Core | Rim | Core | Rim | | A45 | A19 | 205C |
| $SiO_2$ | 37.25 | 36.91 | 37.01 | 37.23 | 37.53 | 37.70 | $SiO_2$ | 34.17 | 34.76 | 35.53 |
| $TiO_2$ | 0.01 | 0.03 | 0.01 | — | — | — | $TiO_2$ | 3.34 | 2.81 | 2.45 |
| $Al_2O_3$ | 20.89 | 20.68 | 21.32 | 21.06 | 21.59 | 21.06 | $Al_2O_3$ | 19.34 | 19.70 | 19.44 |
| FeO | 33.51 | 31.57 | 33.95 | 31.35 | 33.90 | 35.08 | FeO | 21.61 | 21.47 | 20.31 |
| MnO | 4.78 | 7.28 | 3.38 | 7.20 | 2.58 | 2.04 | MnO | 0.29 | 0.28 | 0.08 |
| MgO | 2.82 | 1.91 | 2.89 | 1.96 | 3.54 | 2.67 | MgO | 6.74 | 6.92 | 8.73 |
| CaO | 1.10 | 0.91 | 1.42 | 1.65 | 1.82 | 2.06 | CaO | 0.01 | — | 0.01 |
| $Na_2O$ | 0.01 | 0.04 | 0.05 | — | — | — | $Na_2O$ | 0.09 | 0.21 | 0.22 |
| | | | | | | | $K_2O$ | 9.24 | 9.02 | 8.72 |
| Total | 100.37 | 99.33 | 100.03 | 100.18 | 100.87 | 100.61 | BaO | 0.15 | 0.19 | 0.21 |
| | | | | | | | $H_2O^*$ | 3.87 | 3.90 | 3.95 |
| Si | 6.00 | 6.03 | 5.97 | 6.02 | 5.97 | 6.04 | | | | |
| Al | 0.00 | 0.00 | 0.03 | 0.00 | 0.03 | 0.00 | Total | 98.85 | 99.26 | 99.65 |
| | | | | | | | Si | 5.30 | 5.34 | 5.39 |
| | 6.00 | 6.03 | 6.00 | 6.02 | 6.00 | 6.04 | Al | 2.70 | 2.66 | 2.61 |
| Al | 3.97 | 3.98 | 4.02 | 3.98 | 4.02 | 3.97 | | | | |
| Ti | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | 0.00 | | 8.00 | 8.00 | 8.00 |
| | | | | | | | Al | 0.83 | 0.91 | 0.86 |
| | 3.97 | 3.98 | 4.0Z | 3.98 | 4.02 | 3.97 | Ti | 0.39 | 0.33 | 0.28 |
| Fe | 4.52 | 4.32 | 4.58 | 4.35 | 4.51 | 4.70 | Fe | 2.80 | 2.76 | 2.58 |
| Mg | 0.68 | 0.46 | 0.70 | 0.59 | 0.82 | 0.64 | Mn | 0.04 | 0.04 | 0.01 |
| Mn | 0.65 | 1.01 | 0.46 | 0.78 | 0.35 | 0.28 | Mg | 1.56 | 1.59 | 1.97 |
| Ca | 0.19 | 0.16 | 0.25 | 0.26 | 0.31 | 0.35 | | | | |
| Na | 0.00 | 0.01 | 0.02 | 0.00 | 0.00 | 0.00 | | 5.62 | 5.63 | 5.70 |
| | | | | | | | Ca | 0.00 | 0.00 | 0.00 |
| | 6.04 | 5.96 | 6.01 | 5.98 | 5.99 | 5.97 | Na | 0.03 | 0.06 | 0.06 |
| | | | | | | | K | 1.83 | 1.77 | 1.96 |
| | | | | | | | Ba | 0.01 | 0.01 | 0.01 |
| | | | | | | | | 1.87 | 1.84 | 1.76 |

\* Calculated assuming H = 4.00.

higher Ca and lower Fe. Mg decreases gradually outwards. This pattern is strikingly similar to the residual growth-zoning in the kyanite-zone garnet figured by Anderson and Olimpio (1977, fig. 5). Some other garnets show an increase in Ca near edges, which may be a relic of the same effect (298C, fig. 2a). In 298C and some other rocks from the north-west part of the area, there is also relic 'textural zoning' (cf. Olimpio and Anderson, 1978), small quartz inclusions being found near the centre of garnet.

In most of the garnets (fig. 2b, c), interior zoning is irregular but slight. Thus an interior composition can easily be defined, and compared with matrix plagioclase and biotite compositions. Representative garnet and biotite analyses are given in Table

II. Plagioclase compositions (Table III) were estimated optically or by electron-probe partial analysis for Na, Ca, and K. Biotite and plagioclase compositions are based on the analysis of several grains, chosen at random, from within each sample section. Little variation of either the Fe/Mg ratio of biotite or the Ca content of plagioclase was found. The distribution measures, ln $K_D$(gar–bio) and ln $K$(gar–plag), in Table III are defined by

$$K_D(\text{gar–bio}) = (x_{Fe}^{gar}/x_{Mg}^{gar})/(x_{Fe}^{bio}/x_{Mg}^{bio})$$

and

$$K(\text{gar–plag}) = (x_{Ca}^{gar}/x_{Ca}^{plag})^3.$$
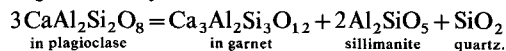
In the unhomogenized regional garnet 572C, the calcium-rich 'outer core' (fig. 2a) was used in the calculation.

TABLE III. *Garnet core compositions and distribution measures*

| Rock | Garnet core | | | | $\ln K_D$(gar-bio) | plagioclase | |
|------|------|------|------|------|--------------------|-----|-------------------|
|      | Alm  | Pyr  | Spe  | Gro  |                    | An  | $\ln K$(gar-plag) |
| A1   | 74.78 | 11.54 | 10.00 | 3.68 | 1.269 | 0.29 | $-6.19$ |
| A2   | 79.88 | 9.70  | 8.11  | 2.31 | 1.337 | 0.23 | $-6.89$ |
| A5   | 75.74 | 8.80  | 11.78 | 3.68 | 1.318 | 0.25 | $-5.75$ |
| A8   | 75.99 | 10.97 | 10.33 | 2.70 | 1.306 | 0.24 | $-6.56$ |
| A45  | 74.80 | 10.79 | 11.24 | 3.16 | 1.309 | 0.25 | $-6.20$ |
| A73  | 74.00 | 12.04 | 11.39 | 2.57 | 1.320 | 0.24 | $-6.70$ |
| A83  | 75.23 | 17.67 | 3.95  | 3.14 | 1.209 | 0.25 | $-6.23$ |
| 270C | 78.05 | 9.68  | 7.96  | 4.30 | 1.292 | 0.25 | $-5.28$ |
| A10  | 70.11 | 8.54  | 15.29 | 6.07 | 1.394 | 0.26 | $-4.37$ |
| A19  | 76.57 | 11.64 | 7.71  | 4.08 | 1.330 | 0.29 | $-5.88$ |
| A32  | 73.86 | 10.37 | 13.16 | 2.61 | 1.279 | 0.19 | $-5.95$ |
| A52  | 67.23 | 10.71 | 18.71 | 3.35 | 1.413 | 0.26 | $-6.15$ |
| A55  | 76.11 | 10.54 | 10.47 | 2.88 | 1.246 | 0.25 | $-6.48$ |
| A72  | 72.90 | 11.63 | 9.92  | 5.54 | 1.320 | 0.25 | $-4.52$ |
| 402C | 75.15 | 10.64 | 9.23  | 4.98 | 1.416 | 0.26 | $-4.96$ |
| A65  | 71.21 | 9.99  | 14.21 | 4.58 | 1.501 | 0.23 | $-4.84$ |
| 205C | 75.33 | 13.67 | 4.12  | 5.19 | 1.440 | 0.30 | $-5.26$ |
| 298C | 77.18 | 10.83 | 9.32  | 2.66 | 1.347 | 0.24 | $-6.60$ |
| 572C | 74.30 | 11.27 | 9.02  | 5.41 | 1.533 | 0.28 | $-4.93$ |
| 614C | 76.42 | 13.43 | 3.87  | 6.28 | 1.327 | 0.37 | $-5.32$ |

In the cordierite–potash feldspar zone, $K_D$(gar–bio) values are closely grouped, though $K$(gar–plag) shows more scatter. Below the cordierite–potash feldspar zone there is scatter in both variables. The data, taken as a whole, define a trend in which the two $K$ measures are correlated (fig. 3). The correlation ($r = 0.560$, $n = 20$) is significant at the 99% level.

*Interpretation.* At equilibrium, the distribution of calcium between garnet and plagioclase should be governed by the reaction

$$3\,\underset{\text{in plagioclase}}{CaAl_2Si_2O_8} = \underset{\text{in garnet}}{Ca_3Al_2Si_3O_{12}} + 2\,\underset{\text{sillimanite}}{Al_2SiO_5} + \underset{\text{quartz.}}{SiO_2}$$

It has been proposed that this equilibrium can be used as a geobarometer (Ghent, 1976), if temperature can be independently estimated, for example from the ion-exchange equilibrium constant $K_D$(gar–bio), which is rather insensitive to pressure (Thompson, 1976; Ferry and Spear, 1978). The isobars and isotherms plotted on fig. 3 are not to be regarded as accurate. In the observed range of $K_D$(gar–bio), the experimental calibration of Ferry and Spear (1978) would give temperatures 40 to 70 °C higher than the empirical calibration by Thompson (1976). The discrepancy may largely be due to the compositional complexity of natural biotites. The barometry is beset by uncertainties in activity-composition relations (Yardley et al., 1980; see below).

The consistent values of $\ln K_D$(gar–bio) from the cordierite–potash feldspar zone, at the high-temperature end of the observed range (fig. 3), are interpreted as indicating that garnet cores, and matrix biotite, in this zone were homogenized with respect to Fe and Mg at high temperatures in the thermal event.

The zoning near garnet edges (fig. 2) is superimposed on the prograde homogenized profiles and must therefore be retrograde (cf. Yardley, 1977). Lower temperatures would be indicated by comparing matrix biotite Fe/Mg with garnet edges than with garnet cores (cf. Tracey et al., 1976). It is possible that such an effect, and marginal Mn enrichment, could be produced by prograde resorption of garnet in favour of a more magnesian mineral, as is argued by Hollister (1977) on an area where garnet is rimmed by cordierite; but there is no petrographic evidence for such an interpretation in the present area, where the marginal zoning affects garnet outside as well as within the cordierite-bearing inner aureole. Nor is there any obvious retrograde resorption reaction, such as replacement by chlorite (cf. de Béthune et al., 1975), and it is possible that the retrograde concentration of Mn in garnet, as well as the Fe, Mg zoning, may be due to ion-exchange reactions (Yardley, 1977).

The correlated variation in $K$(gar–plag) and $K_D$(gar–bio) can be interpreted in terms of two

effects: incomplete prograde reaction, and temperature-dependence of the activity coefficients in the garnet–plagioclase equilibrium. Incomplete reaction would be expected at localities distant from the granodiorite where, indeed, clear evidence is preserved that garnet was not homogenized even by the preceding regional metamorphism (fig. 2a). The residual effect of early regional garnet compositions can be illustrated by considering the inner core of 572C. Calculations of $K$ values from this are physically meaningless, in that it was certainly not in reaction contact with the present matrix assemblage, but carrying out such nominal calculations would give a lower apparent pressure and higher apparent temperature than using the outer core. Partial reaction of such garnets undoubtedly contributes to the irregular

Ca zoning, and to the trend in fig. 3. However, most of the garnet cores are nearly homogeneous, indicating an approach to equilibrium in the aureole, at a nearly constant pressure below that of the regional metamorphism. Their apparently polybaric trend in fig. 3 does not contradict this interpretation, since it may result from changes in the activity coefficients of the Ca components in garnet and plagioclase, as functions of temperature. Yardley et al. (1980) point out that these solid solutions may deviate strongly from ideality with decreasing temperature; this would cause true isobars on a diagram such as fig. 3 to be convex upward. Thus the $P$–$T$ grid on fig. 3 is not a reliable indicator of $P$, and the observed trend is consistent with approach to equilibrium mineral compositions determined almost wholly by $T$. The
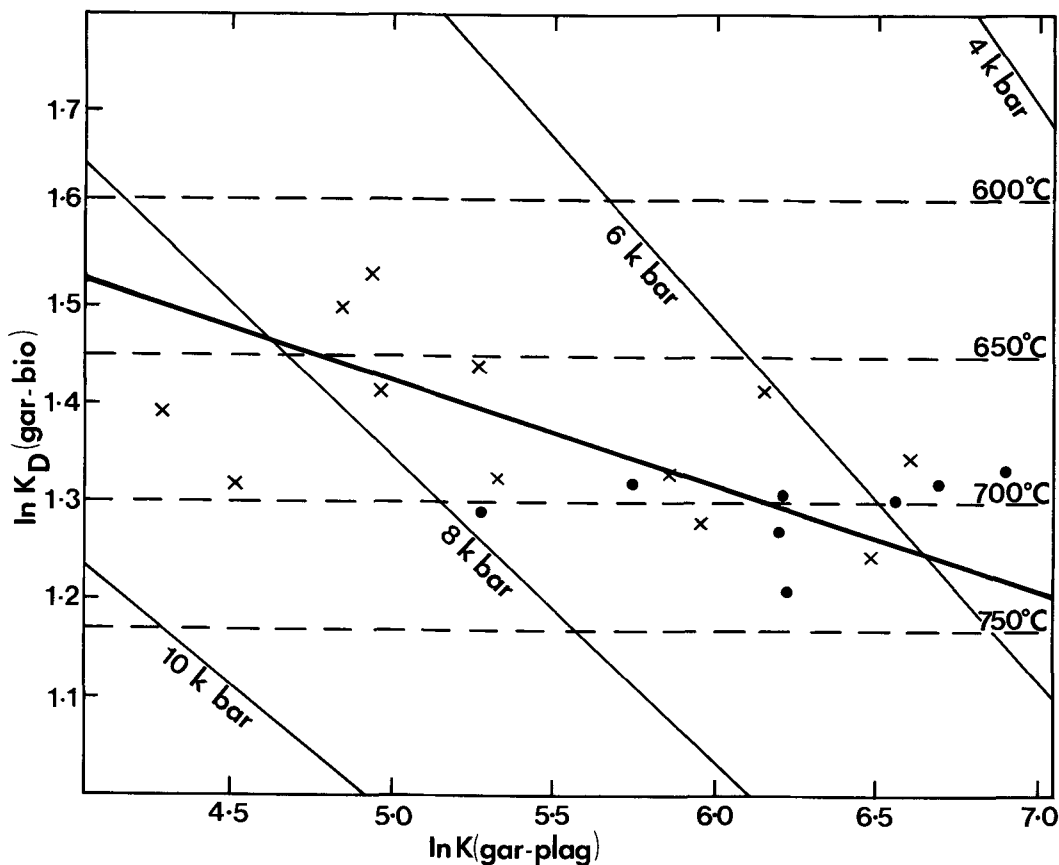


FIG. 3. Distribution of Fe/Mg with biotite and Ca with plagioclase for garnets in sillimanite-bearing assemblages. Circles, cordierite–potash feldspar zone; crosses, other rocks. Estimated equilibrium isotherms and isobars are calculated from the work of Thompson (1976, fig. 1B, assuming pressure-independence), and Schmid et al. (1978) using activity coefficients of 1.276 for An in plagioclase (Orville, 1972) and 0.8 for grossular in garnet (Cressey et al., 1978). Heavy line is least-squares best fit to the data.

absolute $P$ values, from the simple model used in fig. 3, are certainly wrong for the aureole event, being too high. This can be seen from the garnet–cordierite Fe/Mg results of Ashworth and Chinner (1978), whose use is validated by the present inference that garnet was rehomogenized in the cordierite-bearing metamorphic zone. The treatment after Holdaway and Lee (1977) in Ashworth and Chinner (1978) contains a numerical error giving slightly too high estimates of $P$ (and $P_{H_2O}$); the corrected result for the Strontian area is $P = 4.1$ kbar ($P_{H_2O} = 0.5P$), and this is consistent with additional garnet–cordierite data obtained in the present study. Interpreting the trend of $K$(gar-plag) with $T$ in fig. 3 in terms of the effects of non-ideal solid solution on the end-member equilibrium experimentally determined by Schmid et al. (1978), the activity coefficient ratio $\gamma_{Ca}^{plag}/\gamma_{Ca}^{gar}$ should be given by

$$\ln K(\text{gar–plag}) = -15.10 + (3061/T) \\ + (0.6565(P-1)/T) + 3 \ln (\gamma_{Ca}^{plag}/\gamma_{Ca}^{gar})$$

(cf. Yardley et al., 1980, p. 393). Taking $P = 4100$ bar then implies $\gamma_{Ca}^{plag}/\gamma_{Ca}^{gar} \approx 2.7$ at 700 °C and 3.8 at 650 °C, similar to the factors invoked by Yardley et al. (1980) to reconcile their data with the $Al_2SiO_5$ phase diagram.

The data, then, are consistent with a statistical approach to equilibrium except where early zoning is preserved. Below the cordierite–potash feldspar zone, disequilibrium affects some Fe/Mg ratios which should therefore be used with caution. Within the cordierite–potash feldspar zone, partition of Fe and Mg between garnet cores and matrix minerals appears to have reached equilibrium. The garnet–biotite temperatures (fig. 3) from the calibration of Thompson (1976) are consistent with those from the same author's calibration of garnet–cordierite (Ashworth and Chinner, 1978, Table 6). That such methods would not be viable in all areas, even at such high grade, can be seen by interpreting the present observations in terms of literature data for the activation energy, $Q$, for diffusion in garnet. The ratio of diffusion coefficients, $D_2/D_1$, at two temperatures $T_2$ and $T_1$, is given by $D_2/D_1 = \exp[(Q/R)(1/T_1 - 1/T_2)]$ where $R$ is the gas constant. Times required for isothermal homogenization in garnets of the same size, with the same initial compositional variations to provide the driving force for diffusion, are given by $D_1 t_1 = D_2 t_2$ (cf. Yardley, 1977). A conservative estimate of the timescale $t_2$ of the thermal event, relative to the regional timescale $t_1$, can thus be obtained for the Strontian area, by assuming that complete homogenization by the thermal event occurred only in the cordierite–potash feldspar zone (fig. 3). The outer core of 572C is taken to

indicate a lower limit for the temperature at which homogenization was effective in the regional event. It is reasonable to assume the same driving force since the amount of compositional change is similar in the two cases. This approach gives $T_1 \approx 890$ K, $T_2 \approx 960$ K, and $1/T_1 - 1/T_2 \approx 8 \times 10^{-5}$ K$^{-1}$, a difference that is not sensitive to systematic errors of calibration. Then from the published range of estimates for $Q$, 24 to 100 kcal/mole (Yardley, 1977; Freer, 1979), one obtains $t_2/t_1 \approx 10^{-1.1 \pm 0.7}$; the timescale of the thermal event was between a few percent and a few tens of percent of that of the regional metamorphism. This result is highly consistent with the geological evidence that the Strontian intrusion was emplaced at a relatively early stage during regional uplift and cooling (Watson, 1964). The residual regional heat and depth of burial contributed to the retention of the heat of intrusion. The aureole of a later intrusion would have cooled more rapidly. If the timescale of the thermal event was much less than $10^{-2}$ relative to the regional, rehomogenization of garnet would not be expected even at the temperatures of the cordierite–potash feldspar zone. It is presumably in such short-lived thermal events, at shallow depths or after regional temperatures have declined to low values, that new aureole garnets grow with sharp compositional differences from remaining regional garnet (e.g. Edmunds and Atherton, 1971; Okrusch, 1971).

In deep-seated aureoles of the Strontian type, partition of Fe and Mg between garnet cores and matrix minerals will be useful in indicating the metamorphic conditions, but the Ca contents of garnet and plagioclase coexisting with $Al_2SiO_5$ and quartz cannot be interpreted unambiguously until much more is known about the activity-composition relations.

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