Influence of the mode and distribution of garnet and biotite on Grt–Bt thermometry: evidence from a single-sample case study

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Abstract

Detailed microprobe and modal data for a sample of layered, garnetiferous, quartzose paragneiss reveal significant differences in garnet–biotite Mg–Fe distribution coefficients (Kd) — and hence paleotemperatures — determined for leucocratic (modal Grt+Bt<25 vol.%) and mesocratic (Grt+Bt> 25 vol.%) layers. In leucocratic layers, lnKd determined from both the core and rim compositions of minerals shows a range of values that varies sympathetically with the absolute amount of garnet and biotite, and, as demonstrated in other studies, inversely with the distance separating both minerals. Due to the small size (<2 mm) of garnets, which facilitated diffusional re-equilibration during cooling from peak metamorphic temperature, lnKd does not correlate to modal Bt/Grt ratios. The largest garnets, which occur in mesocratic layers, nonetheless tend to preserve the most pronounced (retrograde) zoning patterns (i.e. rimward increase in Fe/Mg), consequently mineral core composition lnKd values correlate with grain diameter except where garnets contain abundant biotite inclusions. The highest Grt-Bt temperatures ($\sim$700°C) are recorded by: (1) the composition of relatively widely-separated (>0.3 mm) grains in highly leucocratic layers; and (2) the core compositions of relatively large (>1 mm), inclusion-free grains in mesocratic layers. More closely spaced garnets and biotites in leucocratic layers, and small grains in mesocratic layers, give a range of temperatures intermediate between T_{max} and diffusional blocking temperatures ($\sim$560°C) recorded by the rim compositions of contiguous grains.

KEYWORDS: garnet, biotite, thermometry, palaeotemperatures, paragneiss.

Introduction

GARNET–BIOTITE thermometry is perhaps the most widely used method of estimating metamorphic paleotemperatures (T). This approach, however, can be compromised by: (1) elevated concentrations of components that can displace the Mg-Fe exchange reaction (e.g. Ti, Al$_{vi}$ in biotite; Mn, Ca in garnet); (2) uncertainties in activity/composition relations used to account for these non-binary components; (3) indeterminate oxidation ratios in biotite; (4) compositional zoning, particularly in garnet; and (5) the susceptibility of biotite and garnet to down-temperature re-equilibration during cooling. As a result, garnet–biotite temperatures tend to be inaccurate and imprecise.

Several studies have addressed these issues. For example, Indares and Martignole (1985a) proposed empirical correction factors to account for the presence of extraneous components in garnet and biotite, thereby extending the use of this thermometer to high-grade rocks. McMullin et al. (1991) and Berman (1990) used linear programming analysis of data from reversed phase equilibrium experiments to determine mixing properties of biotite and quaternary garnet, respectively. Guidotti and Dyar (1991) and Williams and Grambling (1990) noted the role of coexisting oxide phases in buffering the oxidation

ratio of biotite, and Emslie and Stirling (1993) attempted to relate the ferric iron content of biotite to the bulk composition of (igneous) rocks for which both FeO and Fe_2O_3 had been determined. The challenges introduced by mineralogical zoning (e.g. Tracy *et al.*, 1976), and re-equilibration of biotite and garnet compositions to Mg–Fe diffusional blocking temperatures, however, continue to impede the reliable application of this thermometer.

Indares and Martignole (1985*b*) noted that contiguous garnet–biotite pairs in gneissic rocks tend to give lower temperatures than isolated (matrix) grains because their proximity facilitated progress of the exchange reaction during cooling, as reflected by retrograde zoning patterns (i.e. rimward increase in Mn and Fe/Mg) commonly preserved in garnet (e.g. Robinson, 1991, Fig. 44). A corollary of this is that rocks, or parts of rocks, enriched in modal garnet and biotite should yield relatively low Grt–Bt temperatures because the abundance of both phases will ensure numerous mutual grain contacts. Rocks with elevated modal Bt/Grt ratios, however, are expected to preserve biotite and garnet compositions dissimilar to those recording diffusional blocking temperatures because of the 'dilution' of biotite re-equilibration effects (i.e. as each garnet attempts to equilibrate with several nearby biotite grains). Due to mass balance considerations, it is commonly assumed (e.g. Martignole and Pouget, 1993, and references therein) that Bt/Grt ratios exceeding unity are required to ensure that biotite will behave as an infinite reservoir for Fe and Mg, thereby minimizing re-equilibration effects. To our knowledge, this aspect of garnet–biotite equilibria has not yet been quantified in the petrological literature, and is a focus of this contribution.

This study uses microprobe, grain-diameter, and domainal-scale modal- and grain-proximity data to evaluate parameters influencing the compositional re-equilibration of garnet and biotite in layered, quartzose paragneiss from the Grenville Province of southeastern Labrador. To illustrate the effects of fluid flux on Grt–Bt re-equilibration, analytical data were also collected for altered (partly chloritized and saussuritized), biotite- and epidote-rich, mafic gneiss from the same outcrop. Associated metapelitic gneiss from this outcrop, at Barge Bay, contains Grt+Bt+Sil+Pl+Qtz, indicating that maximum metamorphic grade was in the middle amphibolite facies or higher.

The quartzose paragneiss sample was selected for this purpose because layering is defined by a wide range in the proportion and absolute abundance of garnet and biotite; garnet and biotite are the sole Fe–Mg minerals in the rock; garnet, although Fe-rich, approximates a binary composition; and all phases are unaltered. As will be shown, however, the fine grain size of the rock (<2 mm), as well as the distribution of garnet and biotite, influences the results of Grt–Bt thermometry.

Analytical methods

Mineral compositions were determined using a JEOL Superprobe 733 equipped with four wavelength-dispersive spectrometers and one energy-dispersive spectrometer. The operating voltage was 15 kV. The beam current was 12 nA; beam diameter was ~1-2 μm. Count time was 20–40 s. Standards included jadeite (Al, Si, Na), hornblende (Ca, Fe, Mg), sanidine (K), and apatite (P). Probe current was measured with a Faraday cage. The modal composition of the rock in the vicinity of the analysed grains was determined by backscattered-electron (BE) image analysis using the Oxford Link eXL analytical system; BE images were generated over a 256 × 256 pixel area for subdomains examined at a magnification of ×40. Three BE images were taken (and averaged) near each analysed grain (or mineral pair). Core and rim compositions were determined for both contiguous and matrix grains. The analysed grains were selected so as to be representative of the full range of modal layering (as defined by the abundance and relative proportion of garnet and biotite) in the rock. Thin sections were cut perpendicular to modal layering in the gneiss; the sample is not lineated.

Results

The paragneiss contains quartz, biotite, garnet, magnetite, pyrite, and traces of apatite. Collectively, garnet and biotite constitute between ~5 and 70 vol.% of individual layers in the rock, but parts of some mesocratic layers approach 100% garnet+biotite (±magnetite). The sum of modal garnet and biotite (= 'colour index', *sensu lato*) is <25 in what are termed here leucocratic layers (Fig. 1A), and >25 in mesocratic layers (Fig. 1B). Biotite/garnet ratios generally vary between 0.1 and 2 on a domainal scale in the rock, but one mesocratic layer has a Bt/Grt ratio of 7.5. All analysed grains in mesocratic layers constitute contiguous mineral pairs; this is true only for two of the analysed garnet–biotite pairs reported for the leucocratic layers.

Garnets in the quartzose paragneiss are almandine-rich, with subordinate pyrope and minor grossular and spessartine (Table 1). These garnets form idiomorphic to subidiomorphic grains that commonly have inclusion-rich cores. Inclusions consist of minute opaque grains, and biotite. Garnets in mesocratic layers are larger (≤1.5 mm) than those in leucocratic layers (≤0.6 mm). The largest garnets in each layer were analysed, thereby increasing the
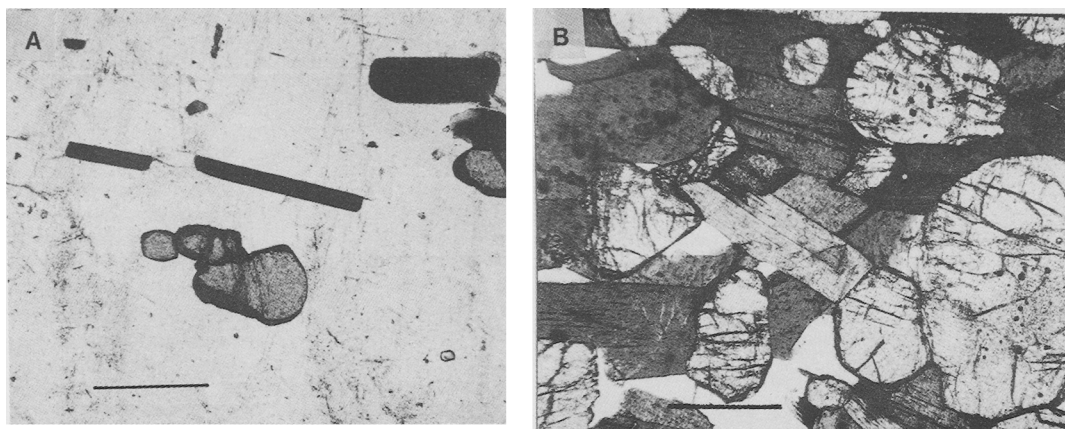
FIG. 1. Photomicrographs of (A) leucocratic and (B) mesocratic layers in garnet+biotite-bearing quartzose paragneiss from the Grenville Province of southeastern Labrador. Plane polarized light. Scale bars = 0.5 mm.

likelihood that measured core and rim compositions are representative of zoning patterns in this mineral.

In most layers, garnet cores have a composition of about $Alm_{88}Prp_7Grs_3Sps_3$. The oxidation ratio, as determined by stoichiometry (Droop, 1987), is low ($X_{Fe^{3+}} \sim 0.02$). Relative to cores, garnet rim compositions tend to be slightly enriched in Fe (and, to a lesser extent, Mn) and depleted in Mg (i.e. they have retrograde zoning patterns), particularly for grains in contact with biotite (Table 1). There are no systematic within-grain variations in grossular concentrations. Although the magnitude of zoning is insignificant in absolute terms, the small amounts of pyrope and spessartine render these components susceptible to large, relative concentration changes across individual porphyroblasts. Consequently, garnets can show a significant rimward increase in Fe/Mg ratios (e.g. from ~ 10 to 15), particularly where in contact with biotite.

Xenomorphic garnets in altered mafic gneiss from the same outcrop are depleted in Fe and Mg, and enriched in Mn, Ca and Fe^{3+} ($X_{Fe^{3+}} \sim 0.18$) relative to garnets in the quartzose paragneiss. They have a composition of about $Alm_{31}Ard_7Prp_4Grs_{37}Sps_{21}$. Metapelite from this outcrop contains subidiomorphic garnets ($Alm_{75}Prp_9Grs_4Sps_{12}$) that show a weak, rimward increase in Mn and a corresponding decrease in Mg.

Biotite occurs as idiomorphic to subidiomorphic crystals, even where occurring as an interstitial phase to garnet in mesocratic layers (Fig. 1B). It is coarser grained in the mesocratic layers (grain area $\leqslant 0.5$ mm^2) than in leucocratic layers (grain area $\leqslant 0.25$ mm^2). The biotite is annite-rich ($X_{Mg} \sim 0.30$), and contains 0.9–2.5 wt.% TiO_2. It is generally unzoned, or only weakly zoned, even where in contact with garnet. Minute biotite inclusions are concentrated in the cores of some of the garnets in mesocratic layers.

TABLE 1. Endmember compositions, modes, and $\ln Kd$ values for garnet and biotite in layered paragneiss from Barge Bay, Labrador

Domain	Type*		Garnet						GS	Biotite			$\ln Kd$
			Diam.	X_{Fe}	X_{Mg}	X_{Ca}	X_{Mn}	Vol. %		Diam.	X_{Mg}	Vol. %	
1	M	c	0.40	0.866	0.054	0.023	0.057	2.21	0.35	0.40	0.175	2.02	1.23
		r		0.874	0.046	0.027	0.054				0.144		1.17
5a	M	c	0.45	0.835	0.077	0.030	0.059	11.11	0.20	0.40	0.312	10.14	1.60
		r		0.845	0.071	0.024	0.060				0.309		1.68
11	C	c	1.00	0.868	0.080	0.025	0.027	36.06	0	0.80	0.295	12.72	1.47
		r		0.880	0.059	0.026	0.035				0.284		1.84
18	C	c	1.20	0.863	0.088	0.026	0.022	56.55	0	0.80	0.268	9.63	1.27
		r		0.881	0.058	0.027	0.033				0.267		1.71

* M = matrix grains; C = contiguous grains; c = mineral core composition; r = rim composition; Diam. = maximum grain diameter (mm); GS = grain spacing (mm).

These biotites tend to be enriched in Ti ($TiO_2 \sim$ 3.0–3.5 wt.%), and are more magnesian ($X_{Mg} \sim$ 0.36) than external grains.

The Fe^{3+} content of biotite is indeterminate from microprobe data, but is likely to be high given the presence of magnetite and absence of ilmenite and graphite in the quartzose paragneiss (cf. Guidotti and Dyar, 1991). Because of this, T estimates reported below are maxima. All layers, however, contain variable amounts of magnetite, and oxidation ratios of coexisting garnet are uniformly low. This suggests, but does not prove, that biotite is saturated in Fe^{3+}, and that its oxidation ratio does not vary substantially. We acknowledge, however, that the Fe^{3+} content of biotite represents as important a variable in Grt–Bt thermometry as some of the other factors evaluated in this paper.

Biotite in the altered mafic rock is relatively magnesian ($X_{Mg} = 0.46$), and unusually enriched in Mn (1 wt.% MnO). The metapelite contains unzoned biotite that is slightly more magnesian ($X_{Mg} \sim 0.37$) and Ti-rich ($TiO_2 \sim 2.7$ wt.%) than that in the groundmass of the layered, quartz-rich gneiss. In addition to garnet and biotite, it contains prismatic sillimanite, retrograde muscovite, and plagioclase (An_{25}).

Evaluation of variables influencing Grt–Bt temperatures

The size and proximity of garnet and biotite, along with their relative abundance, can influence the extent to which these minerals have re-equilibrated during cooling from elevated metamorphic temperatures, and hence the results of Grt–Bt thermometry.
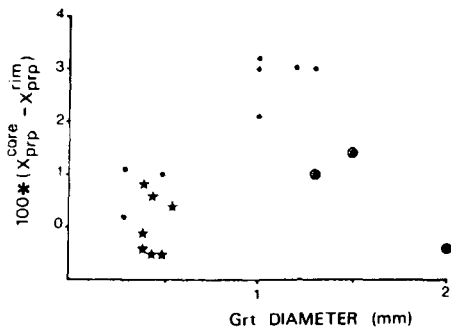

FIG. 2. Variation of garnet core and rim pyrope concentrations with grain diameter. Dots = contiguous mineral pairs (mostly in mesocratic layers; encircled dots = garnets enclosing abundant biotite inclusions; large dot = datum for Mn-rich garnet in altered mafic gneiss); stars = isolated matrix grains (leucocratic layers).

Mineral-pair compositions are reported as the log of distribution coefficients ($\ln Kd = \ln[(FeGrt*MgBt)/(MgGrt*FeBt)]$). Representative $\ln Kd$ values are listed in Table 1.

Grain size potentially plays a dual role in the evaluation of mineral re-equilibration effects. Small grains (particularly of garnet) may be expected to show relatively minor compositional zoning because their large surface area/volume ratio facilitated compositional re-equilibration. Alternatively, they may correspond to the edges of larger grains rather than true cross sections through the mineral. The latter, of course, is avoided by analysing the largest grains observed in thin section.

Spear (1989) evaluated re-equilibration effects in garnet using the Mg–Fe diffusional data of Lasaga *et al.* (1977). He concluded that only the cores of comparatively large (approaching or exceeding 1 cm) garnets are likely to preserve peak T compositions in high grade rocks, i.e. small garnets are more susceptible to pervasive re-equilibration effects during cooling. Notwithstanding the fine grain size of the quartzose paragneiss, there is a positive correlation between grain diameter and the difference in core- and rim- pyrope concentrations ($r = 0.92$, excluding three outliers) in garnets that contact biotite (Fig. 2), i.e. the largest garnets are zoned. This anticipates a relation between $\ln Kd$ and grain size. With the exception of the same three outlying data, there is a strong negative correlation ($r = -0.97$) between garnet diameter and mineral-core $\ln Kd$ values (Fig. 3); there is no such relationship for the mineral-rim $\ln Kd$ data, which vary instead with the proximity of garnet to matrix biotite (Fig. 4). The outliers represent data for relatively large ($\geqslant 1.5$ mm) garnets in the quartzose paragneiss and the altered mafic rock. Two of the outliers (bull's-eye symbol in Fig. 2) represent garnets that contain abundant biotite inclusions. Re-equilibration of the garnet core with these inclusions accounts for the unusually high $\ln Kd$ values associated with these relatively large grains. The third outlier (large dot in Fig. 2) represents the composition of an inclusion-free, xenomorphic, spessartine-rich garnet from the altered, epidote- and andesine-bearing mafic rock. The plagioclase is partly saussuritized, and some biotites are slightly chloritized, indicating fluid infiltration which plausibly promoted retrograde Mg–Fe diffusion in garnet and biotite in this sample.

Although garnets in leucocratic layers in the quartzose paragneiss are uniformly small (~ 0.5 mm), their compositions (when paired with nearby matrix biotites) yield a wide range of $\ln Kd$ values (Fig. 3). In contrast, the rims of contiguous mineral pairs have equilibrated, yielding a narrow range of $\ln Kd$ values. These observations imply a relationship between grain proximity and the degree of composi-
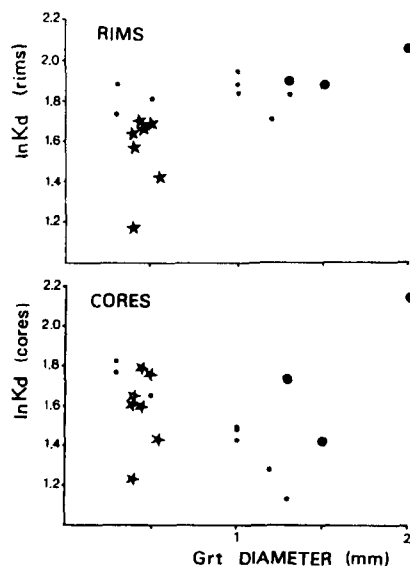
FIG. 3. Variation of $\ln Kd$ with garnet diameter. Symbols as in Fig. 2.

FIG. 5. Variation of $\ln Kd$ determined using mineral rim and core compositions with biotite/garnet ratios. Symbols as in Fig. 2. One point (Bt/Grt = 7.5; $\ln Kd$ (rim) = 1.88, (core) = 1.82) for a mesocratic layer is omitted from each plot.

tional re-equilibration, a feature that has been noted in high-grade rocks by previous workers (e.g. Indares and Martignole, 1985b). Because neighbouring garnets or biotites might have been present in the third dimension, the observation that grains are separated in thin section by relatively wide quartzofeldspathic domains does not ensure that they have avoided deleterious re-equilibration effects. In the present example, grain-proximity data show a strong negative correlation with $\ln Kd$ values ($r = -0.96$ for mineral–rim Kds; $r = -0.99$ for cores) if a single outlying datum is omitted (Fig. 4). Biotite compositions resisted re-equilibration except where very close (within ~ 0.2 mm) to garnet. The outlying datum presumably represents a case where
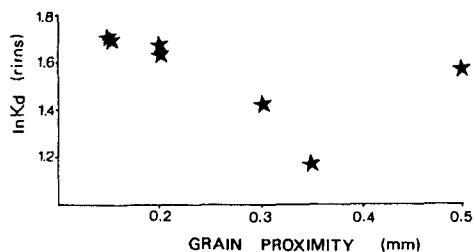

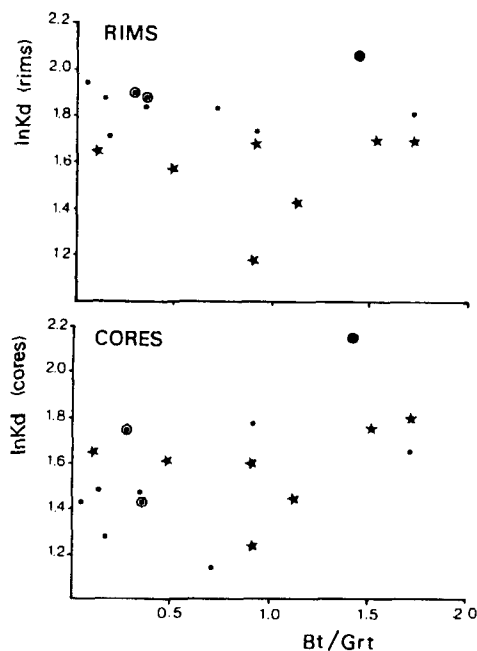FIG. 4. Variation of $\ln Kd$ with grain proximity in leucocratic layers. Symbols as in Fig. 2.

garnet and biotite are relatively distant in two dimensions, but not the third. Nonetheless, the scale of intergrain ionic diffusion implied by these data is small (fraction of a mm) in this rock — a scale comparable to grain size.

The modal abundance (and relative proportion) of garnet and biotite is significant for two reasons. First, garnet and biotite will, by necessity, be relatively closely spaced in mesocratic layers. Second, the modal Bt/Grt ratio may be important because biotite is generally not considered to represent an infinite reservoir for Fe and Mg unless it is more abundant than coexisting ferromagnesian phases (e.g. Martignole and Pouget, 1993).

Garnet/biotite ratios and $\ln Kd$ values should exhibit a negative correlation if abundant biotite served to dilute the effects of down-T re-equilibration of this non-refractory phase. There is, however, no systematic relation between $\ln Kd$ and Bt/Grt ratios (Fig. 5). This may be an artefact of extensive re-equilibration related to the fine grain size of the rock. Coarser-grained samples would be more appropriate to evaluate the effect of Bt/Grt ratios on mineral composition in high-grade rocks. However, due to

difficulties in collecting reproducible modal data for coarse grained rocks, the optimum minimum Bt/Grt ratio required to preserve peak-T biotite compositions would be difficult to determine in samples containing large porphyroblasts.

Interpretation of Grt–Bt temperatures

Values of mineral-rim lnKd for matrix grains are consistently lower than those determined for contiguous mineral pairs. The mineral-rim lnKd data are nearly constant for contiguous grains (Fig. 5), whereas mineral-core lnKd data for the same material show a wide range of values (Fig. 5). This indicates (1) a lack of equilibrium between the cores of contiguous grains (cf. Selverstone and Chamberlain, 1992; Spear and Florence, 1992); (2) the variable degree of re-equilibration of mineral core compositions in mesocratic layers; or, perhaps less likely, (3) that some of these data are not derived from the true cores of the analysed crystals.

In contrast to the cores of contiguous grains, rim compositions equilibrated over a narrow temperature range indicative of the conditions at which effective Mg–Fe diffusion ceased between neighbouring garnet and biotite. The lowest lnKd values determined for garnet–biotite pairs in the leucocratic and mesocratic layers correspond to temperatures of 700°C and 713°C (Perchuk and Lavrent'eva (1983) calibration), respectively. On the one hand, the negative correlation of mineral-core lnKd values with garnet grain size (Fig. 3) suggests that larger porphyroblasts than those reported here should preserve compositions more representative of peak metamorphic T. On the other hand, these represent maximum temperatures because of the indeterminate oxidation ratio of biotite.

Biotites in magnetite-bearing metapelites typically have oxidation ratios ($X_{Fe^{3+}}$) in the order of 0.2 (Guidotti and Dyar, 1991). This lowers Grt–Bt temperatures by several tens of degrees (°C) (e.g. Owen, 1992). This effect, however, can be minor compared with the spurious temperatures derived by mismatching high-T garnets with re-equilibrated biotites. Garnet–biotite temperatures determined from unequilibrated mineral pairs can deviate from the actual peak metamorphic T by hundreds of degrees (cf. Robinson, 1991, Fig. 23). We conclude that, notwithstanding problems introduced by the uncertain oxidation ratios of biotite, re-equilibration effects, and the variables that control them, are an equally if not more important factor than $X_{Fe^{3+}}^{Bt}$. We concede, however, that geologically-valid Grt–Bt temperatures must take into account the possible presence of ferric iron in both minerals.

Contiguous mineral pairs in both mesocratic and leucocratic layers yield diffusional blocking tempera-

tures that average 559°C ($n = 9$). As monitored by lnKd, temperatures associated with mineral core compositions vary with grain proximity and diameter. The quartzose paragneiss therefore preserves on a domainal scale mineral compositions representative of peak metamorphic T and different stages of the subsequent cooling history. Individual T estimates are, of course, meaningful only if they are derived from equilibrated mineral pairs.

The variable re-equilibration effects displayed by the quartzose paragneiss might be more pronounced in rocks containing a relatively feldspathic matrix rather than the quartz-rich matrix that characterizes the layered sample described here. Most ionic diffusion through a quartz-rich matrix will occur along grain boundaries, but it may also occur along cleavage surfaces if (e.g.) feldspar is present, particularly where facilitated by pore fluids. This hypothesis is supported by the extent of re-equilibration of garnet and biotite compositions in the mafic rock and the metapelite, both of which contain plagioclase.

The metapelite is biotite-rich, and contains oligoclase as well as quartz in the groundmass. A unique metamorphic $P–T$ estimate can be determined for this sample from the composition of garnet+sillimanite+quartz+plagioclase (GASP) in conjunction with garnet+biotite. Using mineral rim compositions, updated GE0-CALC (Brown et al., 1988; Berman and Brown, 1988) software (TWQ; Berman 1991) indicates conditions of 545°C at 2.6 kbar. Mineral core compositions, however, indicate conditions of 620°C at 3.9 kbar. Perchuk and Lavrent'eva's (1983) calibration gives very similar Grt–Bt temperatures (mineral rims: 567°C; cores: 606°C). These T estimates are similar to those determined from re-equilibrated garnets and biotites in the quartzose paragneiss.

The metapelite presumably attained the same elevated metamorphic temperature ($\sim$700°C) that was determined for the layered paragneiss. At this temperature, P_{max} (estimated from GASP) is in the order of 5.3 kbar. This is twice the value indicated from mineral rim compositions determined for this sample. In this example, the judicious application of the garnet–biotite thermometer to affiliated rocks provides a basis for interpreting the low pressures determined for the metapelite.

Conclusions

Microprobe data for garnetiferous paragneiss reveal significant differences in Grt–Bt lnKd values determined for leucocratic and mesocratic layers. In the leucocratic layers, where garnet and biotite are small and tend to occur as isolated grains, lnKd shows a range of values that varies sympathetically

with the absolute amount of garnet and biotite, and inversely with the distance between these minerals. Mesocratic layers are relatively coarse grained. Garnet and biotite occur as contiguous grain mosaics, and biotite inclusions occur in some garnets. Excluding inclusion-riddled grains, mineral-core lnKd values are inversely correlated with grain diameter, consistent with the relatively strong compositional zoning preserved by the largest garnets. Maximum Grt–Bt temperatures ($\sim 700°C$) are therefore recorded by the compositions of: (1) grains from layers containing the least amounts of both minerals, which will show a minimal number of mutual grain contacts in three dimensions; and (2) the cores of the coarsest, inclusion-free grains, even though these form contiguous Grt–Bt pairs in mesocratic layers. Mineral rim compositions determined from contiguous grains yield a narrow range of lnKd (and thus T) values approximating diffusional blocking temperatures ($\sim 560°C$). Intermediate temperatures are provided by closely spaced garnets and biotites in leucocratic layers, and small grains in mesocratic layers. Pervasive re-equilibration of the small garnets in the paragneiss hampers the evaluation of the role of domainal-scale modal Bt/Grt ratios in controlling Grt–Bt temperatures determined for this sample.

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