Hierarchy of closure temperatures in granulites and the importance of an intergranular exchange medium (melt?) in controlling maximum Fe-Mg exchange temperatures

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Introduction. Cation exchange closure temperatures are commonly regarded as being controlled by the mineral with the slowest rates of intragranular diffusion of the element(s) of interest (e.g., Lasaga, 1983). Implicit in this view is that volume diffusion within grains rather than intergranular transport between grains is the rate-controlling step in the exchange process. This paper, based on the study of Pattison and Bégin (1994), provides evidence for a hierarchy of closure temperatures in some regional granulites from Quebec. Although several of the cation closures were probably controlled by rates of intragranular diffusion, closure of one very important exchange, Fe-Mg, appears to be dependent on whether or not the garnet and orthopyroxene grains were in contact with other Fe-Mg minerals. In grains isolated by non-ferromagnesian minerals from continued down-temperature Fe-Mg exchange, closure may instead have been induced by loss to the rock of an intergranular exchange medium. Loss of an exchange medium may result in an abrupt transition from effective intergranular communication between minerals on the thin section scale, to ineffective intergranular communication during which exchange between non-touching minerals effectively ceases.

Observational evidence. Compositional maps were prepared of numerous individual garnet and orthopyroxene crystals of varying grain size and in contact with different neighbouring minerals from two garnet + orthopyroxene paragneiss granulites from the regional Minto terrain, Northern Quebec (Bégin and Pattison, 1994). One sample (B69E-90) contains significant amounts of quartzofeldspathic leucosome and is relatively rich in biotite, whereas the second (C10B-89) contains no obvious segregated leucosome and is virtually biotite free, suggesting that it is of higher grade. Plots of composition ((Fe/(Fe + Mg) and X_{Ca} in garnet and Fe/(Fe + Mg) and Al cations in orthopyroxene) versus grain size and neighbouring

minerals revealed the following patterns:

1) core-rim gradients in Fe/(Fe + Mg) in garnet and orthopyroxene are strongly controlled by neighbouring minerals, with strong gradients adjacent to Fe-Mg minerals and negligible gradients against non-Fe-Mg minerals (plagioclase, quartz). The Fe-Mg gradients vary independently from gradients of X_{Ca} in garnet and Al cations in orthopyroxene, the latter two which are generally independent of neighbouring minerals, indicating late localized Fe-Mg exchange after Ca in garnet and Al in orthopyroxene had quenched in.

2) garnet and orthopyroxene grains surrounded only by plagioclase and quartz show generally concentric core-rim gradients in X_{Ca} in garnet (increasing) and Al cations in orthopyroxene (decreasing) but are uniform in Fe/(Fe + Mg), suggesting that the gradients in Ca and Al were quenched in through a relatively high temperature interval during which intragranular Fe-Mg diffusion was fast enough to homogenize gradients in Fe-Mg.

3) cores of garnet and orthopyroxene in contact with other Fe-Mg minerals are progressively more reset in Fe-Mg as grain size decreases. In contrast, cores of even very small grains surrounded only by non-Fe-Mg minerals (plagioclase, quartz) have Fe-Mg compositions close to those of the cores of the largest grains, and are significantly less reset than the cores of larger grain sizes in contact with Fe-Mg neighbours.

These observations suggest the following:

1) Fe-Mg exchange does not retain the high grade part of the rocks' thermal history, and therefore does not retain 'peak' granulite temperatures. Preservation of gradients in Al in orthopyroxene in grains of uniform Fe/(Fe + Mg) suggest that equilibria involving Al solubility in orthopyroxene may provide a better means to obtain the high grade part of the rocks' thermal history (cf. Anovitz, 1991), consistent with crystal chemical considerations.

2) the uniformity of rim Al contents in

orthopyroxene, rim Ca contents in garnet, and Fe/ (Fe + Mg) values of garnet and orthopyroxene grains not affected by late Fe-Mg exchange indicates effective intercrystalline exchange on thin section scale throughout the high-temperature retrograde history of the rocks.

3) the abrupt transition from effective intergranular exchange to strongly localized Fe-Mg exchange adjacent to neighbouring Fe-Mg minerals is most easily explained by loss of an intergranular exchange medium at some point in the cooling hisory of the rocks. Loss of the intergranular exchange medium must have occurred at relatively high temperatures because of the uniformity of Fe/ (Fe + Mg) within and between grains surrounded by non-Fe-Mg minerals (i.e. not subjected to later Fe-Mg exchange).

Calculations. The above zoning patterns were used to guide garnet-orthopyroxene geothermometry calculations, described in detail in Pattison and Bégin (1994). Calibrations used were Fe-Mg exchange (Harley 1984a) and Al-solubility in orthopyroxene with garnet (Harley and Green, 1982; Harley 1984b (the latter corrected by Fitzsimons and Harley, 1994)). These calibrations were chosen because they were derived from the same experiments and thereby provide internal consistency, and because they result in relative temperatures consistent with the zoning patterns.

Using the most refractory core compositions, the maximum Al-solubility temperature for C10B-89 is 890°C compared to a maximum Fe-Mg exchange temperature of 740°C, whereas minimum Al-solubility and Fe-Mg exchange temperatures obtained from mineral rims are 755°C and 540°C, respectively. For B69E-90, the equivalent maximum (core) values are 780°C (Al-solubility) and 680°C (Fe-Mg), and rim values are 680−700°C (Al-solubility) and 480°C (Fe-Mg), respectively. The large difference between the maximum Al-solubility and Fe-Mg exchange temperatures in both samples is consistent with the inferences from the zoning patterns indicating that Fe-Mg continued to exchange and homogenize in the two minerals throughout the interval in which gradients in Al were quenched in. Whereas the Al-solubility temperatures are consistent with (i.e., as high or higher than) minimum temperatures indicated by experimental constraints on orthopyroxene formation (ca. 750−800°C, LeBreton and Thompson, 1988), even the most refractory Fe-Mg temperatures are too low to represent peak temperatures. Notably, the maximum (core) Fe-Mg exchange temperatures in both samples are similar to or lower than the lowest (rim) Al-solubility temperatures, suggesting that, at least in C10B-89, intergranular

Fe-Mg exchange and intragranular Fe-Mg homogenization continued below final closure of Al in orthopyroxene rims, consistent with the zoning patterns.

Loss of an intergranular exchange medium at temperatures at which Fe-Mg was still being homogenized in garnet and orthopyroxene implies that the maximum Fe-Mg exchange temperatures record the temperature at which the medium was lost. Because these temperatures are above the wet granite solidus at the prevailing pressures, the most likely intergranular exchange medium in most rocks is a melt phase rather than a free fluid phase. Melt should be expected to be present in most granulites due to dehydration melting reactions that simultaneously produce melt along with the characteristic anhydrous granulite mineralogy (e.g. Vielzeuf and Clemens, 1987), a view supported by the common occurrence of quartzofeldspathic leucosome in many granulites. Experiments by Watson (1991) indicate that small interconnected melt volumes (< 5 vol. %) are sufficient to increase bulk diffusion rates by orders of magnitude over melt-absent mineral aggregates. Loss of melt by different processes in the two rocks may have induced different maximum Fe-Mg exchange temperatures (which correspond to Fe-Mg closure temperatures in grains isolated from later Fe-Mg exchange): in the virtually biotite- and leucosome-absent C10B-89, high (740°C) Fe-Mg exchange temperatures may indicate relatively high-temperature melt loss, possibly by physical expulsion, whereas the lower (680C) Fe-Mg exchange temperature in B69E-90 may indicate melt loss by freezing, consistent with the biotite- and leucosome-rich nature of the rock. Although melt loss may have induced Fe-Mg closure in grains isolated from later Fe-Mg exchange, Fe-Mg exchange continued to lower temperatures between Fe-Mg grains in physical contact, as evidenced by 480°C Fe-Mg temperatures from rims of touching garnet and orthopyroxene.

Implications for geothermometry of granulites. Fe-Mg exchange thermometry has been the most common means of estimating 'peak' temperatures in granulites. The above results demonstrate that in the regional Minto granulites the most refractory Fe-Mg fractionations (maximum Fe-Mg temperatures) do not record peak temperature conditions, and may instead record the temperature at which an intergranular exchange medium, possibly melt, was lost from the rock. Equilibria involving Al solubility in orthopyroxene appear to provide a better means of obtaining information on the high temperature history of granulites.