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An empirical study of diffusion in garnet

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Abstract

A study of the compositional zoning in garnets that have undergone subsequent metamorphism to staurolite or sillimanite grade confirms the suggestions of some earlier workers that diffusion has taken place in high-grade garnets to eliminate the zoning that developed originally during growth. There is an abrupt change in zoning patterns of garnets within the staurolite–sillimanite transition zone over an approximate temperature range of 50°, with a mean temperature around 640° C. The change in diffusion coefficients with temperature required to eliminate zoning by increased diffusion at the higher grades gives values for Q, the activation energy for diffusion, in the same range as reported values obtained experimentally for comparable diffusion in other silicates. The change in garnet zoning may be used to define an isograd which is not very sensitive to garnet composition or duration of metamorphism. High-grade garnets that have been homogenized by diffusion often show compositional variations at the rims that are attributed to retrograde effects, and it seems probable that at very high temperatures, in excess of 700° C, garnets may be able to adjust composition throughout, during cooling at geological rates.

Introduction

Garnets are among the most commonly zoned minerals in metamorphic rocks, and the correct interpretation of the equilibrium relationships of zoned garnets is essential to the study of garnet-bearing rocks. In general, two types of zoning patterns have been described from almandine-rich garnets. Most commonly, garnets have cores that contain several times as much Mn as the rims, the grossular component may similarly be enriched in the core, and almandine and pyrope contents are greatest at the margins (*e.g.* Hollister, 1966, 1969; Evans, 1966; Atherton and Edmunds, 1966; and many others). This type of zoning is usually attributed to fractionation during growth (Hollister, 1966; Atherton, 1968). In contrast, many high-grade terrains have garnets that are of relatively uniform composition, with zoning commonly restricted to the outer few tens of microns (*e.g.* Blackburn, 1969; Hess, 1971; Grant and Weiblen, 1971). Usually Mn is enriched at the rims, rather than in the core, and the compositional variations are much smaller than those found in lower-grade garnets. The first type of zoning is generally found in low- to moderate-grade rocks, whereas the second type is restricted to high-grade terrains, but it is seldom clear whether the change in zoning pattern is the result of diffusion within preexisting garnet crystals during further heating (Blackburn, 1969; Kretz, 1973), or is due to complete recrystallization of garnet at high grades.

Fractionation models for garnet zoning assume that diffusion in garnet is negligible; however, the development of Mn-enriched rims during corrosion of garnet requires at least limited diffusion (Bethune *et al.*, 1975; Loomis, 1975), and Anderson and Buckley (1973) have developed models for a number of garnet zoning profiles assuming significant diffusion. If the rather uniform composition of high-grade garnets results from the elimination of initial compositional gradients by diffusion, then diffusion must have been effective over distances of millimeters within garnet. Since the rates of self-diffusion of cations in silicates commonly increase exponentially with temperature, it is quite feasible that diffusion may be negligible at moderate grades of metamorphism but becomes significant at slightly higher temperatures.

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Evidence for diffusion in garnet

The basis for this paper is the observed changes in the zoning profiles of garnets from a single pelite horizon in the Dalradian metasediments of Connemara, Ireland, between samples from different metamorphic grades. The metamorphism and deformation of the Connemara Schists in northeast Connemara have been described by Yardley (1976). Garnet grew early in the metamorphic history of the area, generally with a moderate density of inclusions of quartz and ilmenite, sufficient to define an internal fabric that is commonly straight but may be microfolded. Subsequent deformation produced a penetrative regional schistosity that is flattened about the garnets and strongly discordant to the included fabric. Further metamorphism produced staurolite, and in places sillimanite, but the staurolite growth clearly postdates the penetrative schistosity that is wrapped around the garnets, and sillimanite grew still later from staurolite breakdown. The following sequence of metamorphic zones is found in the pelites:

staurolite zone: staurolite + muscovite + quartz + garnet, no sillimanite;

staurolite–sillimanite transition zone: (lower part) staurolite + sillimanite + muscovite + quartz + garnet; (upper part) staurolite + sillimanite + quartz + garnet, virtually no muscovite (garnet is occasionally absent);

higher sillimanite zone: sillimanite + quartz + garnet, no staurolite except as armoured relicts in plagioclase. K-feldspar + sillimanite + quartz + muscovite occur in associated semi-pelites, so this zone is approximately equivalent to the upper sillimanite zone of other workers.

Plagioclase, biotite, ilmenite, and pyrite are found at all grades.

The fabric relationships are believed to demonstrate unequivocally that garnet grains in pelites from all the zones grew early in the metamorphic history of the region. The later development of staurolite and sillimanite, resulting in the present pattern of metamorphic zones, was separated from the growth of garnet by a major phase of penetrative deformation, but the original garnet grains persisted during this subsequent metamorphism. An early fabric, preserved as inclusions in garnet, was entirely obliterated in the rest of the rock prior to the growth of staurolite or sillimanite, and I thus conclude that the garnet grains did not recrystallize during the later metamorphism. Some garnet was corroded during the early stages of staurolite breakdown, but a new growth of garnet is often present in higher sillimanite zone pelites and occurs as overgrowths on cores of early garnet.

Microprobe traverses (Table 1[2]) were made across garnets from all the zones, and examples are illustrated in Figure 1, with sketches of the analyzed grains in Figure 2. All the samples were taken from a single thin pelite layer within an area of about 4 km^2 in the basin of the Failmore River. In all cases the grains studied grew early in the metamorphic history, although sample 210 (Fig. 2D) has well-developed late overgrowths. Figure 1 shows that both the direction and magnitude of the concentration gradients of the R^{2+} cations change in passing from the staurolite zone to the sillimanite zone; Si and Al show no zoning since there are no major substitutions at their sites. Minor irregularities in the zoning profiles generally occur in the proximity of inclusions. Microprobe traverses have been made on 5 additional samples, and core and rim analyses made of garnets from another 6 rocks; the variations in zoning patterns are a simple function of the maximum metamorphic grade that the rocks attained. Since the textures indicate that the garnets did not recrystallize during the maximum-grade metamorphism, I conclude that the differences in composition and zoning pattern between garnets from the different metamorphic zones must result from diffusion of R^{2+} cations within the garnet crystals at high grades, accompanied by exchange of material with the host rock.

Amount of diffusion as a function of grade

Staurolite-zone garnets show little evidence of diffusion if it is accepted that the zoning profiles are those that were generated when the garnet first grew. Minor enrichment in Mn was found in the outer 50 microns of one garnet that had been somewhat corroded. A garnet from Sample 149 (Figs. 1B, 2B), from the lowermost part of the transition zone, has been extensively embayed on one side, while the other is scarcely affected and is mantled by micas. The probe traverse was made across the uncorroded part of the grain and along a 300 micron wide spur towards the former opposite edge of the grain. The Mn content of the grain falls off from the former center and shows enrichment only over the outermost 100 microns; thus the distribution of Mn in the parts of the crystal that remain is similar to that observed in

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[2] To obtain a copy of Table 1, order document no. AM-77-047 from the Business Office, Mineralogical Society of America, 1909 K Street, N.W., Washington, D.C. 20006. Please remit $1.00 in advance for the microfiche.

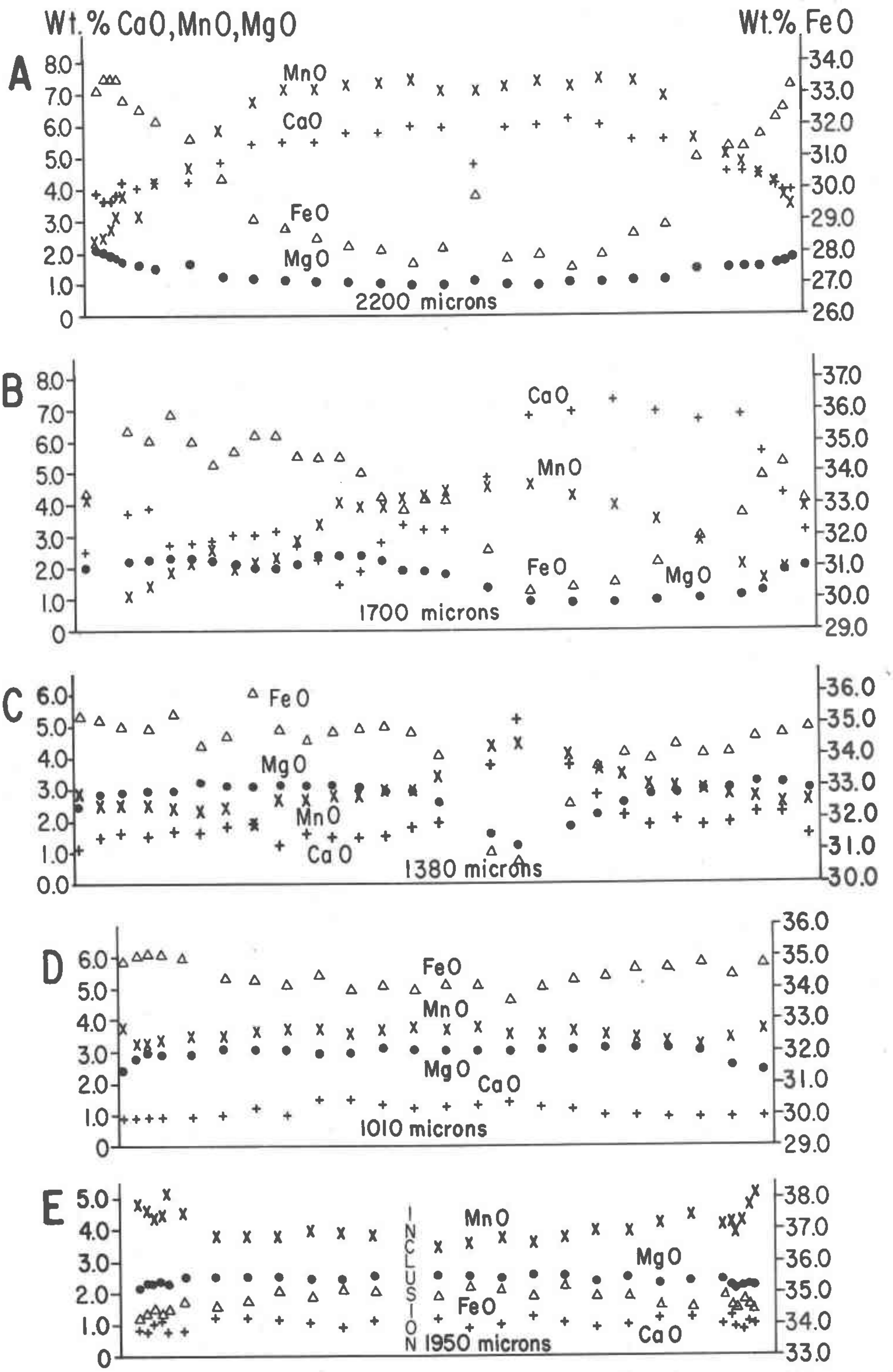

Fig. 1. Representative microprobe traverses of garnets from the Maamturk Pelite Member, Connemara Schists, Ireland. (A) Staurolite zone, Sample 146. (B) Lowermost part of the staurolite–sillimanite transition zone, Sample 149. (C) Upper part of the staurolite–sillimanite transition zone, Sample 111. (D) Higher sillimanite zone, grain with late overgrowth on early core, Sample 210. (E) Higher sillimanite zone, little or no overgrowth, Sample 221. Si, Al, and Ti were also determined at each point, and only point analyses giving totals in the range 97.5–101.5 are plotted. A table of representative analyses is available on microfilm file.

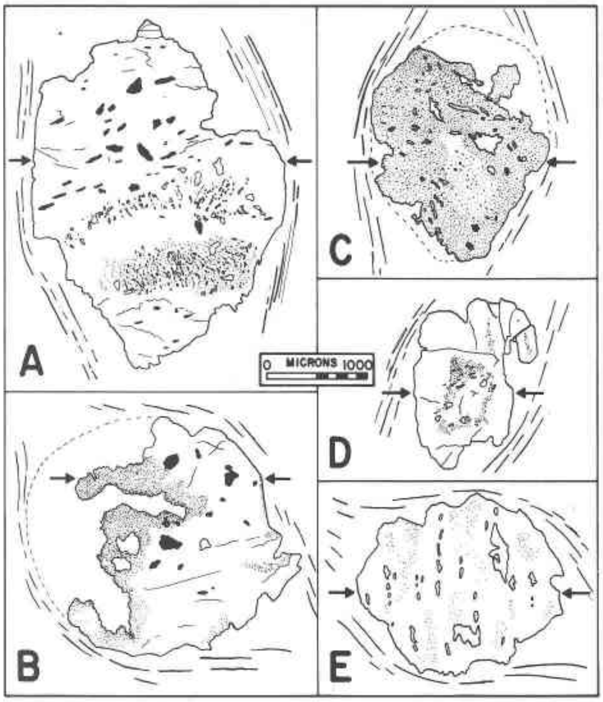

Fig. 2. Sketches of analyzed garnets showing included and external fabrics and location of traverses. Stippling denotes a cloudiness that is developed where diffusion has occurred, except that it is apparently annealed out at the highest grades. Samples are lettered as for Fig. 1. Note that the quartz inclusions in A are concentrated in the hinge regions of small crenulations, opaque inclusions are axial-planar to the crenulations. Dashed lines on B and C denote former extent of grains.

lower staurolite zone garnets. The zoning profiles for Ca, Fe, and Mg are significantly different from those on the uncorroded side. Over the entire length of the relict spur (Fig. 2B) the contents of these elements have tended to equilibrate with the host rock, but the uncorroded part of the grain has retained its original composition; diffusion of these cations has apparently been effective over about 150 microns, so that the entire width of the spur has reequilibrated with the matrix.

Sample 111 (Figs. 1C, 2C) is from the upper part of the transition zone, and some corrosion of the analyzed garnet has taken place. The grain has a small core similar in composition to the staurolite-zone garnets, but the bulk of the crystal is of rather uniform composition and has apparently reequilibrated with the matrix. It is difficult to estimate the depth of penetration by diffusion that this zoning pattern requires since many quartz 'inclusions' may in fact be in communication with the rock matrix, but diffusion has probably been effective over 400–600 microns. Smaller, more corroded, grains from this grade may show no trace of the Mn, Ca-rich core found at lower grades.

Garnets from the higher sillimanite zone have very uniform compositions throughout (Figs. 1D, 1E). In sample 210 (Figs. 1D, 2D) the early garnet cores have well-developed later rims, but there is no marked compositional difference between the two generations of garnet. A slight increase in Ca and Mn towards the core of the grain may be a relict of an original 'normal' zoning pattern (*cf.* Blackburn, 1969). Garnets in sample 221 (Figs. 1E, 2E) are clearly of the earlier generation but have completely adjusted in composition to a high-grade type of zoning profile. This requires diffusion over about 1000 microns.

Many garnets from the staurolite–sillimanite transition zone in Connemara appear cloudy in thin section under plane light. The exact cause of the cloudiness is not known, but it appears to be due to the presence of many very fine cracks, and possibly also fluid inclusions. Opaque precipitates appear to be absent, since even highly-clouded regions give good garnet analyses in the microprobe. The clouded regions correspond to those areas that have changed composition through diffusion, thus in sample 111 (Figs. 1C, 2C), the small core region that remains enriched in Mn and Ca is less clouded than the rest of the grain, and clearly stands out under the optical microscope. In the higher sillimanite zone the cloudiness becomes annealed out, although traces may remain. Rims of new garnet in sample 210 are however perfectly clear. If this phenomenon should prove to be widespread it might be possible to locate garnets in which diffusion had taken place, and measure the extent of the diffusion with the aid of the optical microscope.

In summary, there is a dramatic change in the zoning patterns of early-formed garnets within the staurolite–sillimanite transition zone. Sample 149 occurs just above the sillimanite-in isograd, while Sample 111 is associated with rocks containing quartz + muscovite + sillimanite without K-feldspar. By comparison with the experimental data of Hoschek (1969) and Chatterjee and Johannes (1974) on the appropriate reactions, the temperature range represented by these samples is perhaps around 20–50°, and over this range the depth of penetration by diffusion has increased by about a factor of 2–4. When garnets from the uppermost staurolite zone are compared with those from the higher sillimanite zone, the depth of penetration by diffusion differs by up to an order of magnitude, and the temperature range is 50°–100°.

Although the zoning patterns change dramatically with grade, large changes in the depth of penetration by diffusion are not required. For spherical grains 1 mm in diameter, diffusion to a depth of 150 microns will have little effect on the original zoning profile, but an increase in the depth of penetration by a factor of 4 will completely eliminate the original zonation. The spherical model is not strictly accurate, since many 'inclusions' formed during growth may in fact be in communication with the rock matrix outside the plane of the section. The garnet composition at the edge of quartz inclusions often tends towards that at the outer rim. Thus there will be a tendency to overestimate the amount of diffusion required to modify original garnet compositions.

Comparison with diffusion theory

In order to test whether the abrupt changes in zoning pattern over a small interval of metamorphic grade may reasonably be ascribed to the increasing effectiveness of diffusion with temperature, some calculations have been made of the temperature dependence of diffusion in garnet implied by the proposed interpretation.

The distance over which diffusion is effective in garnet is a function of the diffusion coefficient and the duration of metamorphism. The changes in garnet composition described above can be considered as resulting from the movement of a surface in the garnet, with a composition that is approximately in equilibrium with the rock matrix, towards the core. The distance x of this surface from the edge of the garnet is the depth of penetration. Thus the diffusion in garnet is not in a steady state, and may be described by Fick's second law:

$$\delta C_i/\delta t = \delta/\delta x[D_i(\delta C_i/\delta x)]$$

where C_i is the concentration of the ith component, t is time, and D_i is the diffusion coefficient for component i. The initial stages of the adjustment of the garnet composition by diffusion can be considered as involving exchange between a reservoir of constant composition (the rock matrix) and a uniform garnet core (*cf.* Fig. 1A). Garnets rarely make up more than 4 weight percent of the rock, so small changes in garnet composition do not significantly affect the composition of the matrix. For this situation the solution of Fick's second law for diffusion between a pair of semi-infinite solids is a reasonable approximation:

$$C_i = C'_i[1 + \mathrm{erf}(x/2\sqrt{D_i t})]/2 \qquad (1)\text{(Shewmon, 1963)}$$

This gives the concentration C_i at a distance x from the interface where the concentration of the diffusing species is C'_i in one-half of the couple and zero in the other. This solution is no longer applicable once the core composition begins to undergo significant modification, but it serves to illustrate the general nature of the relationship between the depth of penetration of a surface of some specified composition and D_i and t. For a surface at which i has the concentration C_i to increase its depth of penetration x by a factor n, then the term $D_i t$ must increase by a factor of n^2. It will be assumed that to a first approximation this relationship is true throughout the process of adjustment of garnet composition by diffusion.

If we consider that the staurolite- and sillimanite-zone rocks were raised instantaneously to their maximum temperatures, heated for the same period of time, and quenched, then the difference in $D_i t$ between these two zones would have to be entirely in D_i. However, it would be more reasonable to suppose that heating and cooling were not instantaneous and to substitute the integral:

$$\int_{t=0}^{t=t} D_i\, dt \qquad (2)$$

for the $D_i t$ term, where D_i varies exponentially as a function of $1/T$ according to the Arrhenius relationship:

$$D_i = D_0 e^{-Q/RT} \qquad (3)$$

D_0 and Q are constants for diffusion of a particular species in a particular host, Q being the activation energy for diffusion, R is the gas constant, and T the absolute temperature.

Because the regional heating and cooling rates are not known, it will be assumed that heating and cooling were instantaneous and that rocks of all grades were metamorphosed for the same period of time. It then becomes possible to calculate a value for Q without knowing the absolute values of the diffusion coefficients, by using the relationship:

$$\mathrm{d}\ln D_i/\mathrm{d}\,1/T = -Q/R \qquad (4)\ \text{(from equation 3)}$$

The calculation is not very sensitive to the assumption of instantaneous heating and cooling.

In the previous section it was estimated that x had increased by a factor of 3 over a temperature range of 30° or by a factor of 10 over 75°, corresponding to increases in D of 9 and 100 respectively (from equation 1). The mean temperature is around 640°. Substituting in *4* we get values of Q of approximately 70 and 100 kcal per mole respectively; this crude calculation allows comparison of the temperature dependence of the diffusion coefficients required to produce the observed changes in garnet composition by the postulated model, with the experimentally-determined temperature dependence of cation diffusion in other silicates. Foland (1974) measured the values of Q for diffusion of Na, K, and Rb in orthoclase under hydrothermal conditions, and obtained values in the range 53–73 kcal per mole. Misener (1974) reported values of Q between 50 and 65 kcal per mole for interdiffusion of Fe and Mg in olivine, depending on the bulk composition of the grain and direction of movement. These values are in general agreement with the empirical estimates for diffusion in garnet. It appears to be quite reasonable for the zoning profiles of garnets to change abruptly over a narrow temperature interval as the result of diffusion, as inferred from the textural evidence and microprobe analyses.

The use of garnet zoning to define an isograd

Because the change in garnet zoning takes place abruptly over a small temperature interval in this area, it may provide a useful isograd. In the examples described, changes in garnet composition accompany elimination of zoning by diffusion, and thus diffusion is accompanied by exchange of material with the matrix. However, as long as diffusion within the garnet grains is the slowest step in any reaction in which the garnet may be involved, diffusion will become effective in all almandine garnets at the same temperature, irrespective of the mineralogy of the matrix and any reactions. For this reason the "change-in-zoning-pattern" isograd would be a useful way of tracing a thermal surface through rocks in which more diagnostic assemblages are lacking, and of comparing the temperature of metamorphism of garnet-bearing rocks with otherwise dissimilar assemblages.

The principal factors that might vary the temperature at which growth zoning profiles are eliminated are variations in garnet size and composition, in the duration of metamorphism, and in pressure.

The effects of grain size are the most readily corrected for since relicts of earlier zoning patterns may persist in large grains after smaller crystals have homogenized. In the previous section it was assumed that diffusion rates of all the principal R^{2+} cations in garnet were similar. Clearly this is not the case (*e.g.* Fig. 1B), and garnets enriched in the more mobile cations may be able to homogenize more rapidly than others. We can consider the diffusion as involving exchange of Mn^{2+}, Mg^{2+}, Fe^{2+}, and Ca^{2+} through the garnet lattice, subject to the single constraint of electric neutrality. Thus diffusion of the fastest cation will be constrained by the flux of one or more of the other cations in the opposite direction, while diffusion of the slowest cation will not be affected by the rates of movement of the others. Hence the temperature at which the original zoning of the slowest cation is eliminated will be independent of garnet composition, except in so far as the diffusion coefficients themselves depend on the local bulk composition of the garnet. No information is available on this point, but many garnets in non-calcareous metasediments at the appropriate grades are of rather similar composition.

Equation 1 shows that variations in the duration of metamorphism will have the same effect on the depth of penetration by diffusion as equivalent variations in D. Thus for diffusion to be as effective in staurolite-zone rocks as it was in the Connemara sillimanite zone, the duration of metamorphism would have to be increased by a factor of perhaps 100. Minor variations in the duration of metamorphism or rate of heating and cooling along a single belt will have a negligible effect on the temperature at which the pattern of garnet zoning changes, but there might be significant differences when very different types of terrain are compared. The mean temperature of the zone over which the profiles change might differ by as much as 100° between a thermal aureole and an ancient basement terrain.

The effect of pressure on diffusion in solids depends on the detailed diffusion mechanism, but commonly tends to reduce D (Shewmon, 1963). This has been documented by Misener (1974) for olivine. It seems doubtful that normal pressure variations within the crust would have a very significant effect on diffusion in a very rigid mineral such as garnet. The presence of water may have a large effect on diffusion in silicates (Griggs and Blacic, 1965), but most garnets in amphibolite-facies metasediments must have formed in a hydrous environment. I therefore conclude that changes in duration of metamorphism, and possibly in garnet composition, are likely to be the principal sources of any variations in the temperature at which growth zoning is eliminated from garnets.

Interpretations of garnet compositions in equilibrium studies

In many studies it is assumed that the rim composition of the garnet was in equilibrium with the adjacent phases at the peak of metamorphism. The present study suggests that this is not the case at high grades. If the original growth zoning was eliminated from sillimanite-zone garnets by diffusion at the peak of metamorphism, then the marginal zonation, commonly enrichment of Mn at the rim, cannot have been present at that time and must have been imposed subsequently during cooling as proposed by Grant and Weiblen (1971). Hess (1971) has also ascribed the development of Mn-rich rims about homogeneous garnets to a retrograde event. Thus in the study of garnets from high-grade rocks with compositional profiles similar to these illustrated in Figures 1D–1E, the core composition should be taken as representing the high-grade equilibrium composition. The width of marginal zonation may be a measure of cooling rate. Often, the development of Mn-rich margins is associated with breakdown of the garnet, and Loomis (1975) has presented some detailed models for this process. In other cases there is no sign of corrosion where Mn enrichment has occurred; for example, Dudley (1969) has figured perfectly euhedral garnets with Mn-rich rims, and thus the Mn enrichment may also result from cation-exchange reactions alone.

An important implication of the strong temperature-dependence of diffusion in garnet noted here is that at still higher temperatures, above 700° C, diffusion may become so effective in garnet that grains are able to adjust composition throughout during initial cooling. Thus cation-exchange thermometry may give anomalously low results at very high grades, except under exceptional circumstances of rapid cooling.

Conclusions

Self-diffusion of R^{2+} cations in almandine garnet becomes sufficiently rapid to eliminate compositional zoning formed during growth over a narrow temperature range between the first appearance of sillimanite in staurolite schists and the first breakdown of muscovite + quartz in the Connemara region. The temperature range over which the zoning is eliminated appears to be about 50° for 1–2 mm diameter grains, and the mean temperature is about 640° C ± 30°. The change in zoning is sufficiently abrupt and regular to mark an isograd that should be independent of the assemblage in which the garnets occur. The temperature at which diffusion becomes effective is relatively insensitive to the duration of metamorphism, but will vary by a few tens of degrees between terrains that were metamorphosed for markedly different periods of time. Garnets homogenized by diffusion commonly have margins enriched in Mn, and this is interpreted as a retrograde phenomenon; hence the core composition of such grains represents the high-temperature equilibrium.

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