

CALCIUM ZONING IN ALMANDINE GARNET, WISSAHICKON FORMATION, PHILADELPHIA, PENNSYLVANIA

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ABSTRACT

Garnets with clearly developed Ca zoning generally are associated with zoned plagioclase in samples of interlayered pelites and calcareous sandstones in the Wissahickon Formation northwest of Philadelphia, Pa. Systematic Ca zoning is absent in metamorphic assemblages with high variance, although the garnet may be zoned in other elements. Apparently calcium zoning in metamorphic almandine-rich garnets develops (1) during metamorphic reactions involving plagioclase-garnet equilibria; (2) as a result of reactions between other minerals in low-variance assemblages; and (3) in volumes of rock subject to changes in calcium content due to diffusion during metamorphic recrystallization. Where several of these mechanisms for producing Ca zoning operate simultaneously, the zoning profiles are complex. Where the zoning is relatively simple, the trends of the calcium zoning in almandine, when considered in conjunction with the zoning in the coexisting plagioclase, can be used in the interpretation of the thermal history of the sample; the calcium zoning is independent of the mechanisms which produce zoning of Mn, Fe and Mg.

SOMMAIRE

Les grenats contenus dans les échantillons de pelites et de grès calcaires interstratifiés de la formation Wissahickon au nord-ouest de Philadelphie, Pa., États-Unis, et montrant une zonation Ca bien développée, sont habituellement associés au plagioclase zoné. Une zonation systématique du calcium est absente dans les associations métamorphiques à variance élevée, bien que le grenat puisse être zoné pour d'autres éléments. Apparemment, la zonation de calcium dans les grenats métamorphiques riches en almandin se développe (1) pendant les réactions métamorphiques impliquant un équilibre entre le plagioclase et le grenat, (2) comme résultat des réactions entre d'autres minéraux en associations à faible variance, et (3) dans des volumes de roches soumises à des variations du contenu en calcium en raison de la diffusion pendant la recrystallisation métamorphique. Les profils de zonation sont très complexes quand plusieurs de ces mécanismes de zonation du calcium agissent au même endroit. On utilise la zonation du calcium dans l'almandin de pair avec la zonation du plagioclase coexistant, lorsque celle-ci est assez simple, afin d'établir l'histoire thermique de cet échantillon. La zonation du Ca est indépendante des mécanismes produisant la zonation du Mn, du Fe et du Mg.

(Traduit par la Rédaction)

INTRODUCTION

As our knowledge of the details of garnet chemistry in metamorphic rocks continues to grow it is apparent that the calcium content of almandine garnets may be used as a geothermometer or a geobarometer in certain metamorphic assemblages (Ghent 1976; Hensen *et al.* 1975). In addition, the calcium zoning patterns in garnets usually differ, often markedly, from the zoning trends shown by Fe, Mg and Mn (Crawford 1974; de Béthune *et al.* 1968; Linthout & Westra 1968). Most of the detailed studies of garnet equilibria have focused on the Fe, Mg and Mn zoning and the chemical interactions between garnet and other ferromagnesian phases. Ca zoning has been described, but little attention has been paid to the mineral parageneses and possible reactions which govern the variation of the calcium in almandine-rich garnet.

An advantage to studying compositional variation within refractory minerals such as garnet is that zoning patterns, where preserved, provide information on the development of the mineral assemblage from the moment when those minerals first nucleate in the rocks. The thermal histories of most metamorphic rocks can be placed in one of three general categories: rocks which were subjected to rapid heating with considerable overstepping of mineral reactions; rocks in which the mineral assemblages have been affected by extensive retrograde reactions as the rocks cooled, and those which recrystallized under prograde, near-equilibrium conditions and do not show evidence of retrograde effects. The types of calcium zoning in garnets observed in the first case were discussed by Crawford (1974) and in the second by de Béthune *et al.* (1975) and Hollister (1976). This paper deals with garnets in rocks of various compositions from the staurolite zone of the amphibolite facies of a prograde metamorphic terrain.

The samples are from the Wissahickon Formation exposed along Wissahickon Creek in northwestern Philadelphia, Pa. Most of the section consists of alternating pelitic and moderately psammitic units several tens of meters thick in the exposed section. Locally, thin bands (2-

10 cm) of calcareous sandstone are interlayered in the pelitic units. The samples were collected from two outcrops of pelite and calcareous sandstone with the intention of comparing the garnet compositions in rocks of different calcium contents.

Two sets of samples were studied in detail, one collected slightly above the staurolite isograd and the other midway between the first appearance of staurolite and the isograd representing the reaction staurolite+quartz=garnet+kyanite. The samples were about 1 km apart, perpendicular to the strike of the isograds. In this area the exposed width of the staurolite zone is 5.5 km. No differences were found in mineral composition between the two localities. As the assemblages are in the staurolite stability field, metamorphic temperatures during recrystallization are inferred to be between 550° and 650°C (Fig. 1). Within the calcareous layers muscovite oc-

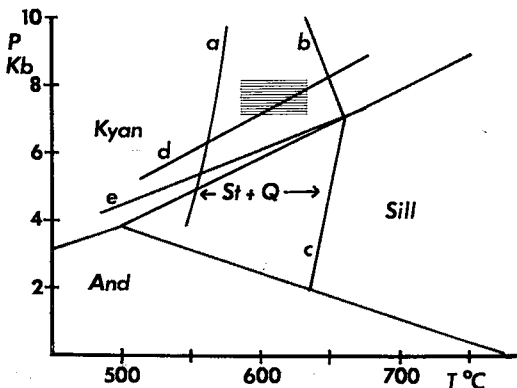


FIG. 1. Pressure-temperature field for samples studied. Experimentally determined equilibrium curves: (a) chlorite+muscovite=staurolite+biotite+quartz+H₂O (Hoschek 1969); (b) staurolite+quartz=garnet+kyanite+H₂O and (c) staurolite+quartz=garnet+sillimanite+H₂O (Ganguly 1972); (d) muscovite+zoisite+quartz=anorthite+K-feldspar+H₂O (Johannes and Orville 1972; Hewitt 1973); (e) muscovite+zoisite+quartz=anorthite+K-feldspar (Ackermann & Karl 1972). Aluminosilicate stability curves from Holdaway (1971).

curs with clinozoisite, plagioclase and quartz; potassium feldspar was not seen. The breakdown of clinozoisite+muscovite+quartz approximately parallels the kyanite=sillimanite isograd (Ackermann & Karl 1972). Data from Johannes & Orville (1972) and Hewitt (1973) place this reaction about 1 kbar higher than suggested by Ackermann & Karl (Fig. 1). The experimental data need to be adjusted to account for 10-15 mole-percent albite, in the plagioclase and 10-15

mole percent pistacite in the clinozoisite. However, these substitutions will shift the equilibrium in opposite directions and probably will tend to cancel each other. In higher grade rocks in this area, staurolite breaks down in the stability field of kyanite. This suggests a pressure of at least 7 kbar for the region, which is compatible with the observed stability of the clinozoisite+muscovite+quartz assemblage.

One fundamental observation which is of considerable importance in deciphering the calcium zoning in almandine is the common association of zoned plagioclase with calcium-zoned garnet. Thus the calcium content of a metamorphic almandine garnet is controlled by plagioclase-garnet equilibria according to reactions such as: grossular+Al₂SiO₅+quartz=anorthite. In addition, equilibria involving additional calcium-bearing phases may be involved: zoisite+quartz=anorthite+grossular+H₂O, and anorthite+calcite+quartz=grossular+CO₂. To a first approximation therefore, the grossular content of the garnet will reflect the Na/Ca ratio of the rocks, and secondly the metamorphic history of the sample, which influences the reactions between Ca-bearing phases.

ANALYTICAL PROCEDURES

In each specimen the garnets were analyzed for Mg, Fe, Mn, Ca, Al and Si with an ARL EMX electron probe microanalyzer. The analyses were made along traverses through the center of the grains at 6 and 10 micron intervals

TABLE 1. COMPOSITION OF GARNETS ILLUSTRATED IN FIGURES 2-4

	SiO ₂	Al ₂ O ₃	FeO*	MnO	MgO	CaO	Total
1a core	37.4	21.1	27.3	10.0	2.4	3.2	101.4
1a rim	37.4	21.3	32.5	3.9	3.8	2.6	101.5
1b core	37.4	20.8	29.6	6.5	3.6	3.0	100.9
1b rim	37.5	21.2	31.5	5.4	3.9	2.2	101.7
2 core	36.5	20.7	36.7	.2	1.5	4.6	100.2
2 rim	36.9	21.1	35.4	.2	3.5	3.0	100.2
3 core	37.3	21.1	26.7	3.3	2.3	9.7	100.5
3 rim	38.1	21.3	25.3	2.0	2.6	11.1	100.4
4 core	37.6	21.5	25.5	5.8	2.1	9.2	101.2
4 rim	37.6	21.1	30.0	2.2	2.6	7.0	100.5
5 core	38.0	21.1	33.0	2.5	1.8	5.3	101.7
5 rim	38.5	21.1	31.1	1.5	3.3	5.5	101.0

Numbers of ions on the basis of 12(O)

	Si	Al	Fe	Mn	Mg	Ca
1a core	2.98	1.99	1.82	.68	.28	.27
1a rim	2.97	1.99	2.15	.26	.45	.22
1b core	2.98	1.96	1.98	.44	.43	.26
1b rim	2.98	1.99	2.09	.37	.40	.19
2 core	2.96	1.98	2.49	.01	.18	.40
2 rim	2.96	1.99	2.37	.02	.41	.26
3 core	2.97	1.98	1.78	.22	.27	.83
3 rim	3.00	1.97	1.67	.14	.31	.94
4 core	2.96	1.99	1.68	.39	.25	.78
4 rim	2.99	1.98	2.00	.15	.31	.60
5 core	3.01	1.97	2.19	.17	.21	.45
5 rim	3.03	1.96	2.05	.10	.38	.47

*Total Fe reported as FeO

(depending on the size of the grain). Similar traverses were made across zoned plagioclase, although here the stepping interval was 4 microns. The analyses were corrected using a modified Bence-Albee correction procedure. The diagrams are plotted in such a way as to smooth out statistical counting fluctuations and to show only actual variations in the apparent compositions of the minerals. Non-statistical errors in the analyses contribute, in part, to the irregularities of the zoning profiles. However, whenever checks were run by analyzing the same traverse more than once, most of these fluctuations were reproduced.

LOW-CALCIUM ASSEMBLAGES

The semi-pelitic samples studied are mineralogically simple, and hence possess a high variance

(>2). Also, they contain minerals which are stable through a wide range of metamorphic conditions: quartz, plagioclase, biotite, muscovite, and minor garnet. The calcium content of the garnet shows no systematic zoning pattern within single grains and remains constant between garnets in adjacent layers in contrast to the behavior of Fe, Mg and Mn (Table 1, Fig. 2). This reflects an equilibrium composition for the plagioclase and garnet across a thin-section despite variations in the absolute and relative abundance of other elements. Examination of the calcium content and zonation in almandine reported by other investigators also shows that calcium content is not sensitive to variations in abundance of other divalent cations in garnet (e.g. Crawford 1974; de Béthune *et al.* 1975; Linthout & Westra 1968).

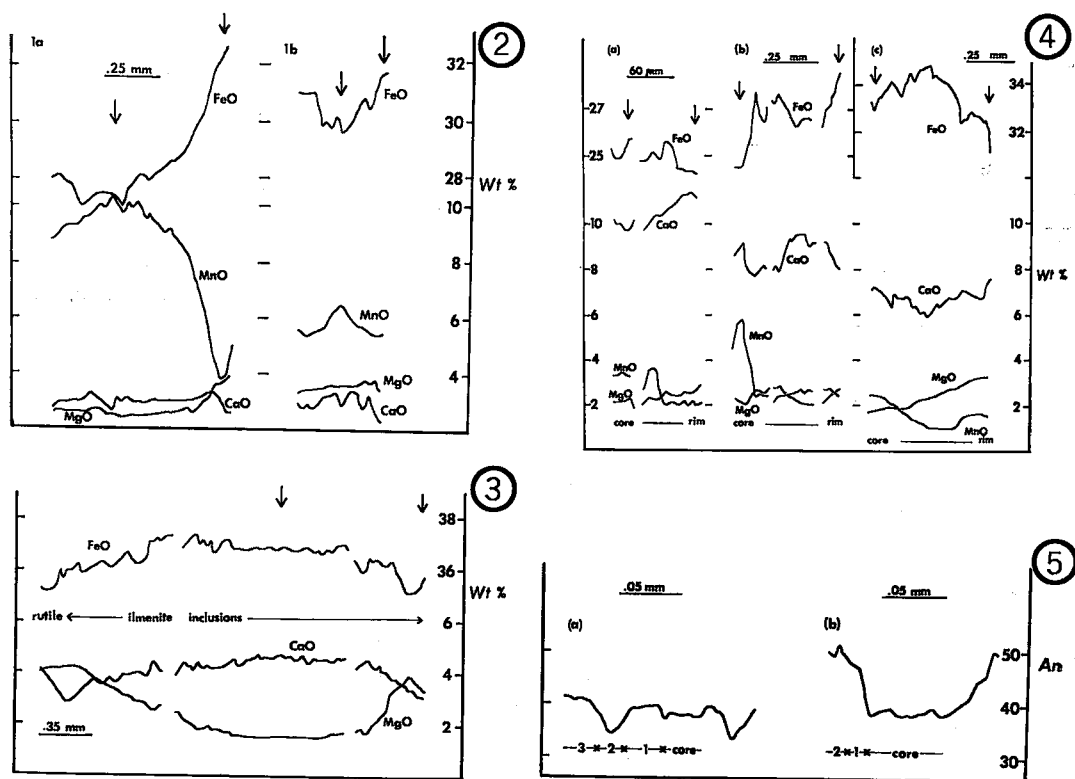


FIG. 2. Zoning profiles across garnet 1a and 1b from a semipelitic schist. Chemical analyses for points indicated by arrows reported in Table 1.

FIG. 3. Zoning profile across garnet 2 from a pelitic schist. Arrows as in Figure 2.

FIG. 4. Zoning profiles from core to rim of three garnets: (a) in the center of a calcareous sandstone layer, (b) in the reaction band between the calcareous layer and the pelite and (c) adjacent to the pelite. Arrows as in Figure 2.

FIG. 5. Zoning profiles across two plagioclase grains: (a) in the reaction band between the calcareous layer and the pelite, (b) adjacent to the pelite.

Aluminous samples from the Wissahickon Formation at this metamorphic grade commonly contain too many phases for divariant equilibrium when judged by phase-rule criteria: kyanite-staurolite-garnet-biotite-muscovite-quartz-plagioclase-ilmenite-rutile. This type of situation is not unusual and has been explained by postulating additional components such as Zn in staurolite and Mn in garnet to increase the variance. Garnet 2, illustrated in Figure 3, is from this type of sample. As can be seen from the analysis in Table 1, the Mn content of garnet is negligible; the same holds for ilmenite, and the staurolite is free of Zn. Alternatively, the low variance may indicate that the fugacity of H_2O is buffered by the mineral assemblage. There is no petrographic reason to suppose $P_{H_2O} < P_{total}$ for the schists in question. In particular, the calcareous layers within the pelites all contain clinozoisite. Clinozoisite will form only in assemblages with a high mole fraction of H_2O , so the pelites have acted as a reservoir of H_2O , diluting the CO_2 produced in the calcareous layers by decarbonation reactions. Graphite and sulfide minerals are absent from these pelitic schists, precluding any suggestion of significant C or S components in the fluid phase.

The opaque inclusions in garnet show that the garnet nucleated and grew while ilmenite was stable in the matrix. Staurolite and kyanite, on the other hand, contain only rutile inclusions and rutile grains are found in the outermost rim of the garnet and in the matrix. Matrix rutile is, in turn, rimmed by ilmenite. The mineral relationships suggest that most of the garnet growth preceded the nucleation of staurolite and kyanite. When staurolite appeared, ilmenite was replaced by rutile. Thus the low variance probably reflects a lack of simultaneous equilibrium among all of the phases in the sample.

Zoning in garnet 2 reflects the effects of the changing mineral equilibria as the P and T conditions affecting the sample changed. This is a direct result of the low variance of the mineral assemblage. Figure 3 shows a gradual decrease in Ca and Fe and an increase in Mg during most of the growth history of the garnet. At the point where the inclusions change from ilmenite to rutile there is an inflection in the zoning profiles and the Ca trend reverses whereas the Mg trend flattens out. The inflection in the zoning pattern apparently coincides with a change in the mineral assemblage due to a discontinuous reaction during prograde metamorphism. Trzcinski (1971 and pers. comm.) has shown in similar low-variance assemblages that plagioclase is involved in reactions which produce and

consume staurolite. His observations suggest the reaction muscovite+chlorite+calcic plagioclase+ilmenite = staurolite+biotite+calcic garnet+rutile+quartz+sodic plagioclase+ H_2O . Supporting this postulated reaction are observations that Ca increases in the rim of garnet 2 and that, at the same point as the inflection in the Ca profile, the inclusions in the garnet change from ilmenite to rutile. The reaction consumes plagioclase and it is interesting to note that the plagioclase with garnet 2 is sparse and corroded. It is not possible to further document this reaction in the Philadelphia region due to a horst of older gneisses which interrupts the metamorphic zonation in the Wissahickon schist below the staurolite isograd. Further west, along Brandywine Creek where lower grade rocks do occur, chlorite is abundant in rocks below the staurolite isograd and biotite becomes an important phase only above that isograd.

CALCAREOUS ASSEMBLAGES

Calcareous sandstone layers within the Wissahickon Formation at this grade show a reaction zone which separates them from adjacent more pelitic layers. The centers of the calcareous layers consist of clinozoisite, quartz, plagioclase and garnet. Some layers also contain muscovite; others are hornblende-bearing. These layers are separated from pelitic schist by a 1-5 mm band consisting of quartz, plagioclase, and minor garnet. Within this band some of the garnet contains clinozoisite and, occasionally, hornblende inclusions. The inclusions show the calcareous sandstone - pelite interface band formed, at least in part, from a mineral assemblage resembling the one in the interior of the calcareous layers. Subsequent reactions, however, have simplified the mineralogy of this band relative to that of the adjacent calcareous layer. The mechanism of formation of bands between pelitic and calcareous layers in schists has been described by Vidale & Hewitt (1973).

In the centers of the calcareous layers where clinozoisite occurs both as inclusions in garnet and in the matrix, both the garnet (Fig. 4a) and the plagioclase show an increase in Ca content from core to rim. Adjacent to the pelite, in those bands with no clinozoisite in the garnet or the matrix, the zoning pattern shows Ca enrichment in the core and rim separated by a Ca-depleted zone (Fig. 4c).

In contrast to the unzoned or simple Ca zoning pattern observed in garnet in the semipelitic and pelitic schists, the garnets and the associated plagioclases in the reaction band between the calcareous layer and the pelite show fairly complex Ca zoning. Here (Fig. 4b) clinozoisite in-

clusions occur in a high-Ca garnet core. The core is rimmed in turn by low-, high- and low-Ca zones. It is possible to predict which garnets will have the most complex variation in Ca as the adjacent plagioclase shows oscillatory zoning consisting of at least four zones (2 high-Ca, 2 low-Ca, Fig. 5a).

Despite the variable complexity of Ca zoning, zoning of Mg and Mn shows uniform patterns similar to those of the garnets in the semipelitic and pelitic schist regardless of the position of the garnets in the rock. Mn is highest in the center and decreases outward. The characteristic bell-shaped curve of the Mn zoning in the core of the grain flattens and may show a slight increase at the margin (4b, 4c). Inclusions in the garnets are concentrated in that part of the grain showing the strongest Mn zoning. This is interpreted as a reflection of most rapid growth and simultaneous rapid depletion of Mn from the matrix for the central part of the garnet with numerous inclusions.

The Mg content increases from core to rim whereas the Fe profile is more irregular. Where Ca is strongly zoned it is clear that Fe zoning mirrors Ca zoning. Fe zoning is also affected by the changes in Mn and Mg. It is not useful to discuss Fe/Mg ratios without recognizing the effect, on the Fe content, of Ca as well as Mn. Mg values, however, do not seem to show effects due to Ca zoning; the regular increase in Mg contents may reflect garnet growth during increasing temperature conditions.

The mineralogical and chemical variations observed in calc-silicate layers embedded in schist can be explained, in part, by diffusion of calcium down its activity gradient. This kind of diffusion of non-volatile components, and its effects, has been described by Vidale & Hewitt (1973) and Thompson (1975). The presence of an actively diffusing species will reduce the number of inert components in the region between the pelite and the center of the calcareous layer. This will decrease the number of coexisting phases, as seen in the interface band. Outward diffusion of calcium during mineral growth will also produce a gradient in the composition of the plagioclase and the garnet, as is observed. The overall calcium content of garnet and plagioclase is highest in the centers of the calcareous layers and decreases as the contact with the pelite is approached. The variation in composition of individual grains will also reflect the changing Ca content in the matrix during mineral growth.

Interpretation of the zoning pattern in the calcareous layers relies on the assumption that the zoning in garnet and plagioclase grains repre-

sents a growth history that reflects changing metamorphic conditions during crystallization. A second assumption is that successive zones can be correlated between plagioclase and immediately adjacent garnet, counting inward from the outermost parts of the grains. This assumption is supported by the observation that the number of zones found in each mineral is in almost every case the same. Finally, if the zoning in plagioclase and garnet follow the same trend (both increasing or both decreasing in Ca content) it is assumed that zoning is controlled by the overall availability of calcium to the growing minerals. On the other hand, if the zoning patterns are antithetic, the most reasonable explanation is that the two minerals are reacting in exchange equilibrium. The slope of curves on a *P-T* diagram representing equilibrium between grossular and anorthite are positive (Boettcher 1970). An increasing temperature or decreasing pressure during crystallization will therefore result in more calcic plagioclase in equilibrium with less calcic garnet for any plagioclase-grossular reaction. The reverse will be true as the minerals equilibrate during retrograde metamorphism. Of course, both changes in overall Ca content of the local system and exchange reactions between garnet and plagioclase may operate simultaneously and produce very complex relations.

In those parts of the samples studied in which plagioclase and garnet grew in equilibrium with clinozoisite (center parts of calcareous layers and garnets with clinozoisite inclusions in the cores) both plagioclase and garnet are uniform in composition (Fig. 4a). Ca released to the system by the breakdown of clinozoisite shows as a slight increase in the Ca content of both minerals. Adjacent to the pelite, the core compositions of garnet and plagioclase are uniform but are surrounded by a zone of less calcic garnet and of more calcic plagioclase (Fig. 4c, 5b). As clinozoisite is not present, the assemblage is probably not buffered with respect to calcium. The zoning presumably reflects a temperature increase, for the plagioclase becomes more calcic and, in the garnet, the Ca content decreases. A pressure decrease would produce the same zoning pattern. However in the adjacent pelite the evidence, cited above, for growth of garnet, followed by staurolite and kyanite, supports a model of mineral growth under increasing temperature conditions. In this part of the sample the outer rims of both minerals are enriched in calcium possibly due to clinozoisite breakdown in the calcic central part of the layer and outward diffusion of the Ca ions toward the pelite.

In samples from the mid-staurolite zone, the

distinct interface band between the calcareous and pelitic layers is most well-developed. Here the zoning pattern of garnet and plagioclase becomes more complex; the number of composition zones in both minerals increases (Fig. 4b, 5a). Interpreting the growth history of the minerals becomes subjective except that they have progressed through the same thermal history (increasing T) as the garnet in the immediately adjacent pelite, discussed above, and the garnets in the more calcareous part of the sandstone layers. In this part of the rock in which diffusion of Ca, as well as other species, was most active, as shown by the simplified mineralogy, the minerals grow in an ever-changing chemical system. The combination of this effect and possible exchange reactions among the calcic minerals as they grow could be expected to produce the complex zoning patterns that appear.

DISCUSSION

The origin of zoning in garnet may be attributed to one or more of four processes:

(a) Preferential fractionation of one element into garnet during growth to produce a depletion of that element in the matrix and hence a depletion in later garnet zones. This type of zoning occurs only if the element is preferentially incorporated in garnet relative to all other minerals. For calcium it might occur at low metamorphic grade, where plagioclase is still albite or if the Ca content of the rock is very low and the growth of both garnet and plagioclase removes the bulk of the Ca from the reacting system. This will produce decreasing Ca zoning profiles in both plagioclase and garnet and was not observed in this study.

(b) Zoning due to exchange reactions as changes in temperature and pressure change the values of partition coefficients for a pair of minerals. There is no other common mineral in metamorphic rocks in which Ca substitutes freely for Fe, Mg, and Mn, hence the grossular component of garnet does not enter into this type of reaction. This mechanism of zoning, which is of great importance for Fe and Mg zoning patterns, does not apply to Ca.

(c) Zoning due to continuous and discontinuous reactions. The analysis above suggests that this mechanism provides the reason for most Ca-zoning in garnet and plagioclase of the Wissahickon Formation. Continuous reaction relations which involve plagioclase and garnet on opposite sides of the reaction produce antithetic zoning patterns in the two minerals. As temperature increases or pressure decreases, the plagioclase becomes more calcic and the garnet less so. The reverse is true as temperature decreases or pres-

sure increases. Discontinuous reactions, such as breakdown of clinozoisite, will provide added Ca to the system and cause the calcium content of both phases to increase. Discontinuous reactions which do not primarily involve calcic minerals, such as reaction to form of staurolite, may also produce a distinctive change in the composition of the calcic phases. This will occur in low-variance assemblages in which the compositions of all minerals are fixed for each P and T . Finally, if the chemical gradient is steep enough, diffusion will cause chemical changes, mirrored by changes in calcium content, in garnet and plagioclase removed from the immediate vicinity of the reaction involved in providing the Ca ions.

CONCLUSIONS

The analysis of zoning of calcium in these garnets from rocks with similar thermal histories but variable compositions shows that such zoning patterns are best deciphered when related to zoning in coexisting plagioclase. The occurrence of a similar number of different zones in those two Ca-bearing minerals permits the assumption that the zones can be correlated. In high-variance and constant-composition assemblages there is little or no systematic zoning in the calcium content of garnet and coexisting plagioclase. However, in samples with low variance, or samples in which there has been a significant change in chemistry due to diffusion during prograde metamorphism, calcium zoning in garnet and coexisting plagioclase is evident. A model for the development of zoning as a consequence of continuous and discontinuous reactions during near-equilibrium prograde metamorphic reactions can explain the zoning patterns observed and provides a mechanism for inferring thermal history of the sample during the growth of the garnet and plagioclase.

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