

Garnet composition and zoning in the determination of temperature and pressure of metamorphism, central Massachusetts

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Abstract

The distribution of Fe, Mg, Ca, and Mn within individual zoned garnets of medium- to high-grade metamorphosed pelitic rocks from west-central Massachusetts has been studied in detail with an automated electron microprobe, and is consistent with variations predicted for the Fe–Mg–Mn continuous reactions taking place at each metamorphic grade. This regular behavior allows use of compositions of garnet and coexisting phases for geothermometry and geobarometry if *P-T-X* relations of appropriate continuous reactions can be calibrated. Preliminary calibrations of garnet–biotite and garnet–cordierite Fe–Mg exchange reactions and several Fe–Mg–Mn continuous reactions have been used to evaluate *P-T* conditions of metamorphism in central Massachusetts. Results indicate pressure in the range of 5 to 7 kbar and a west to east temperature gradient from about 580–605°C for kyanite–staurolite grade to 650–700°C for sillimanite–orthoclase–cordierite grade. Pressure and temperature estimates are consistent with regional tectonic reconstructions indicating synmetamorphic uplift in the Bronson Hill anticlinorium and downbuckling in the Merrimack synclinorium.

Introduction

The Bronson Hill anticlinorium and Merrimack synclinorium in west-central Massachusetts, east of the Mesozoic border fault, are composed of late Precambrian and/or Cambro-Ordovician basement gneisses mantled by metamorphosed Middle Ordovician, Silurian, and Lower Devonian sedimentary and volcanic rocks, and various syntectonic plutons (Robinson, 1967a, 1967b; J. B. Thompson *et al.*, 1968; Field, 1975). Zones of Acadian (Devonian) regional metamorphism have been mapped in pelitic schists of the Partridge, Littleton, and Erving Formations (Robinson, 1963, 1967a, b; Hess, 1969; Hall, 1970; Field, 1975) which indicate increasing metamorphic grade from west to east over a distance of about 50 km (Fig. 1). Assemblages characterizing the zones and the probable reactions separating them are summarized in Table 1. Locations and estimated

modes of specimens studied in detail are listed in Table 2.

Various continuous (multivariant) reactions characterizing these zones have been studied in detail (Robinson, 1963; Hall, 1970; Tracy, 1975). Recent attempts to calibrate continuous reactions in terms of pressure (*P*), temperature (*T*), and activity of H₂O (*a*H₂O) (A. B. Thompson, 1976a, b) use experimentally determined and calculated pure-Fe and pure-Mg end-member reactions and Fe–Mg exchange potentials calculated from distribution coefficient (*K_D*) data. These calibrations are here applied to detailed petrographic and microprobe information on pelitic rocks, in an attempt to understand the pressure–temperature history of this portion of the Appalachian mountain belt.

The near ubiquity of garnet in assemblages in pelitic rocks over a wide range of metamorphic grade and bulk compositions makes it a useful mineral for continuous geothermometry and geobarometry. The nature and origin of the complex chemical zoning of

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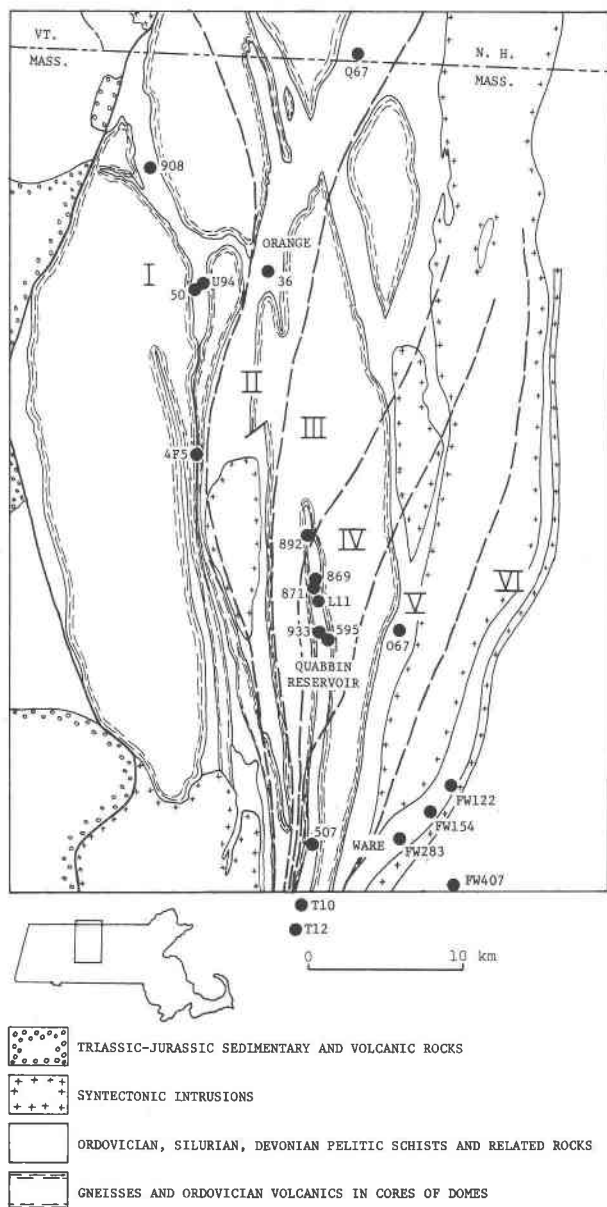


FIG. 1. Generalized geologic map of west-central Massachusetts showing the distribution of metamorphic zones and locations of specimens.

garnet must be understood, however, before compositions may be used for pressure and temperature determinations. The garnets from different metamorphic zones show element distribution patterns that are complex functions of rock bulk composition, specific continuous reactions in which garnet is involved, P - T history of the rock, homogeneous diffusion rates within garnet, and possibly also the availability of metamorphic fluids at the various stages of garnet development.

Assemblages, continuous reactions, and garnet zoning

During microprobe investigation of the samples, it was noted that garnets from different metamorphic zones exhibit a variety of element zoning patterns. It appears that slow homogeneous diffusion rates within garnet, unlike other phases, permit preservation of growth zoning at medium grades of metamorphism. This means that element distribution patterns within garnets may yield information on the nature of prograde and retrograde continuous reactions involving garnet. Differing styles of garnet zoning may be discussed conveniently in a framework of increasing metamorphic grade, as reflected by the various metamorphic zones observed in central Massachusetts.

Zones I and II

Metamorphic Zone I is characterized by assemblages of Kya-Mus-Sta-Gar-Bio with Qtz + Gph + Ilm \pm Pla \pm Pyh (Table 1), though kyanite is restricted to rocks with magnesian bulk compositions (Robinson, 1963; Hall, 1970). Zone II differs from Zone I only in the presence of sillimanite in place of kyanite. Sillimanite is much more abundant in this zone than kyanite in Zone I. Garnets from both zones are usually euhedral and generally contain inclusions of quartz and ilmenite, in some cases showing evidence of rotation during growth. These garnets exhibit "normal" chemical zoning (Hollister, 1966) characterized by Ca- and Mn-richer cores and Fe- and Mg-richer rims. Single garnets from these zones have been examined in great detail with an automated electron microprobe, and chemical contour maps of a typical garnet in sample 908 from Zone I are presented in Figure 2. These represent the two-dimensional distribution of end-member components in a section cut approximately through the center of a grain.

Zone I samples 908 and 4F5 show a roughly concentric and regular zoning with the notable exception of a ridge of higher pyrope content near the edge, caused by a considerable drop in Mg/Fe at the edge. Representative analyses of Zone I (908, 4F5) and Zone II (36Y, Q67) garnets have been plotted in terms of atomic percent Fe, Mg, and Mn in Figures 3a and 3b. In Zone I samples (Fig. 3a) initial outward zoning shows a decrease in Mn accompanied by a slight increase in Mg/Fe, consistent with the predicted trend of garnet compositions involved in the continuous Fe-Mg-Mn reaction



TABLE 1. Metamorphic zones, mineral assemblages, and abbreviations

Zone	Assemblage	Discontinuous Reaction
I	Qtz-Bio-Gar-Sta-Kya-Mus+Pla-Gph-Ilm+Pyh Qtz-Bio-Gar-Sta-Mus+Pla-Gph-Ilm+Pyh	
		Kya = Sil
II	Qtz-Bio-Gar-Sta-Sil-Mus+Pla-Gph-Ilm+Pyh	
		Sta + Mus + Qtz = Sil + Gar + Bio + H ₂ O
III	Qtz-Bio-Gar-Sil-Mus-Pla-Gph-Ilm+Pyh	
IV	Qtz-Bio-Gar-Sil-Mus-Ksp-Pla-Gph-Ilm+Pyh	Mus + Pla + Qtz = Sil + Ksp + H ₂ O
V	Qtz-Bio-Gar-Sil-Ksp-Pla-Gph-Ilm+Pyh	
		Bio + Sil + Qtz = Gar + Crd + Ksp + H ₂ O
VI	Qtz-Bio-Gar-Crd-Sil-Ksp-Pla-Gph-Ilm+Pyh	

Mineral Abbreviations

Bio - Biotite	Ksp - K-feldspar	Pyh - Pyrrhotite
Crd - Cordierite	Kya - Kyanite	Qtz - Quartz
Gar - Garnet	Mus - Muscovite	Sil - Sillimanite
Gph - Graphite	Pla - Plagioclase	Sta - Staurolite
Ilm - Ilmenite		

with increasing temperature (A. B. Thompson, 1976a). From element distribution in the limiting binary systems it is predicted that $T_{Mg} > T_{Fe} > T_{Mn}$, where T_{Mg} represents the temperature of the reaction in the pure-Mg system at constant P and a_{H_2O} . The Zone II garnets show a similar trend toward lower Mn, but have very little, if any, change in Mg/Fe from core to rim. This cannot be explained by a continuous Fe-Mg reaction, but may represent the effect of Mn on the discontinuous Fe-Mg reaction



which can be thought of as a continuous Fe-Mg-Mn reaction with $T_{Fe} \geq T_{Mg} > T_{Mn}$.

The abrupt change in zoning toward lower Mg/Fe observed near the margins of Zone I garnets (Figs. 2 and 3a) cannot be easily related to prograde continuous Fe-Mg-Mn reactions, and may be related to continuous reactions involving other types of cation exchange or to retrograde reactions, on the basis of features observed in higher-grade garnets (see below).

Zones III, IV, and V

Garnets from Zone III (Mus-Sil), Zone IV (Mus-Sil-Ksp), and Zone V (Sil-Ksp) are distinct from those of Zones I and II in that they are rarely euhedral and contain few inclusions. As indicated in Table 1, Zones II and III are separated by reaction

TABLE 2. Estimated modes and locations of specimens

	Qtz	Pla	Mus	Ksp	Kya	Sil	Bio	Sta	Gar	Crd	Ilm	Gph	Pyh	Fm*	Qu**	Ref.†
908	63	tr	#20			10	tr	5			tr	1		L	N	(1)
50Y	50	tr	17		tr	26	4	1			1	1		E	M	(1)
U94X	53	tr	19			15	7	2			1	3		L	M	(1)
4F5AY	45	tr	25		3	16	2	7			1	1		P	M	(1)
36Y	68		16			2	12	tr	1		tr	1		P	O	(1)
Q67X	50	2	16			6	23	tr	3		tr	tr		L	G	(1)
892U	34	18	3			7	36		tr		tr	1	1	P	Q	(2)
871	39	7	9	9		6	29		tr		tr	tr	1	P	Q	(2)
869	25	10	5	6		19	32	1			tr	1	1	P	Q	(2)
L11Y	25	25		5	5	34		5			tr	tr	1	P	Q	(2)
933A	30	10		5	14	30		7			1	2	1	P	Q	(2)
933B	15	10		tr	10	43		20			tr	1	1	P	Q	(2)
595C	28	26	2	5	5	31		tr			tr	1	2	P	WD	(2)
507B	29	11		8	8	38		2			tr	3	1	P	WD	(2)
T10B	25	8		10	18	34		3			tr	1	1	P	Pa	(2)
T12A	30	10		13	12	29		5			tr	1		P	Pa	(2)
067D	37	10		5	10	27		10			tr	1		L	Pe	(3)
FW283	25	tr		30	15	15		12	1	2	tr			P	Wa	(4)
FW154	15			5	23	15		20	20	2	tr			L	Wa	(4)
FW122	11	1		20	20	35		10	2	tr	1			L	Wa	(4)
FW407	30	13		12	12	15		8	2	tr	tr	8		P	Wn	(4)

*Formation: P= Partridge, L= Littleton, E= Erving.

**Quadrangle: N= Northfield, M= Millers Falls, O= Orange, G= Mount Grace, Q= Quabbin Reservoir, WD= Winsor Dam, Pa= Palmer, Pe= Petersham, Wa= Ware, Wn= Warren.

†References: (1) Robinson (1963), Hall (1970); (2) Tracy (1975); (3) Robinson, unpublished data; (4) Field (1975).

‡tr -Trace, less than 1 percent.

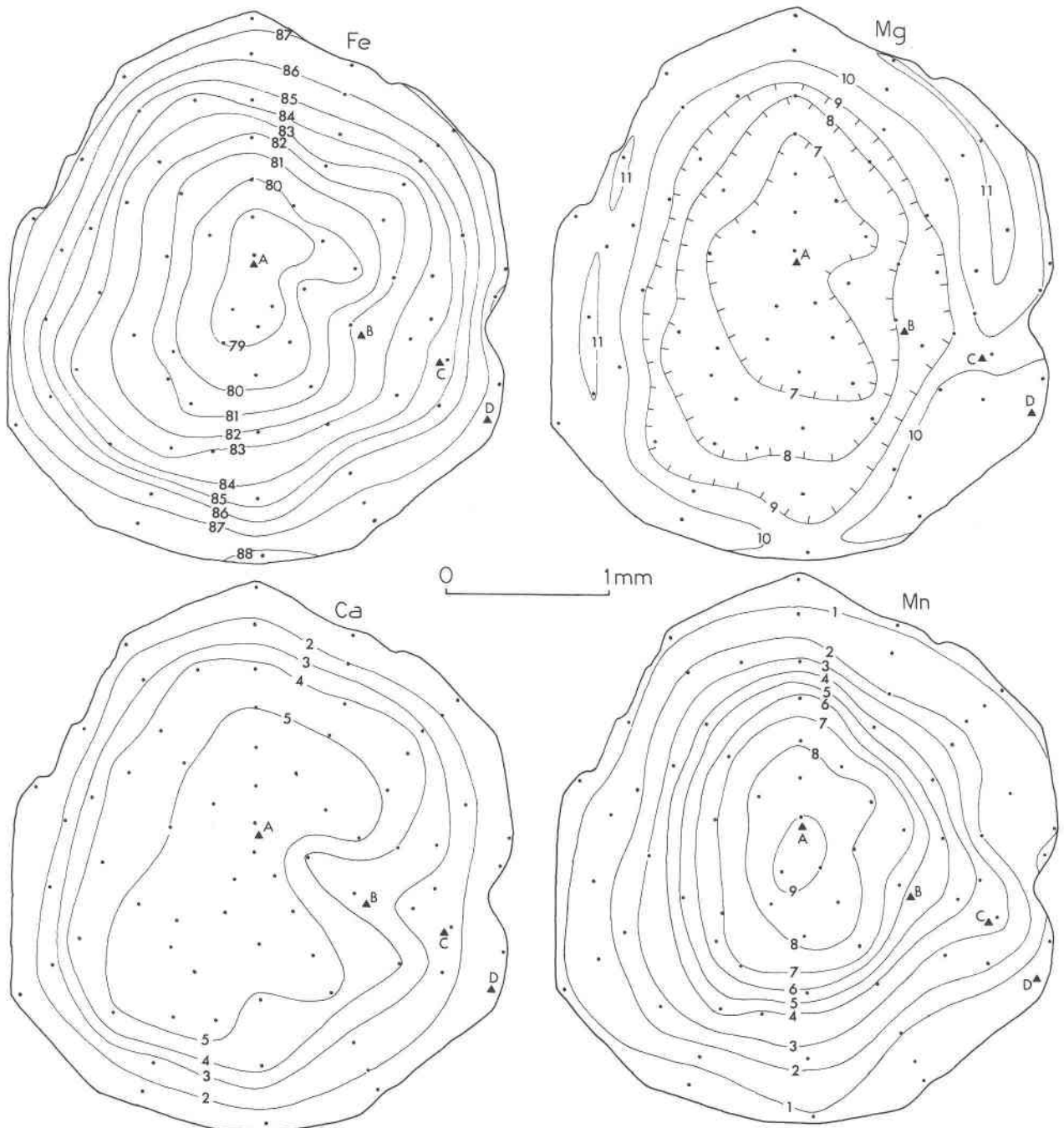


FIG. 2. Contours of atomic percent Fe, Mg, Ca, and Mn in a garnet from specimen 908, Zone I. Dots indicate microprobe analysis spots, and triangles indicate analyses of ilmenite inclusions, keyed by letter to Table 4. Probe section was specially prepared to pass as close as possible to the center of a subhedral crystal.

(2), which marks the upper stability of staurolite in these pelitic rocks. Figure 3b illustrates the zoning trend for a Zone III garnet, 892U. The rim has lower Mn than the core, but also has considerably lower Mg/Fe. The only prograde continuous Fe–Mg–Mn reaction which can operate in Zone III,



cannot be used to explain garnet growth in this zone, since garnet is *consumed* by this reaction, for which $T_{\text{Mn}} > T_{\text{Fe}} > T_{\text{Mg}}$. An alternative reaction producing garnet involves ilmenite,

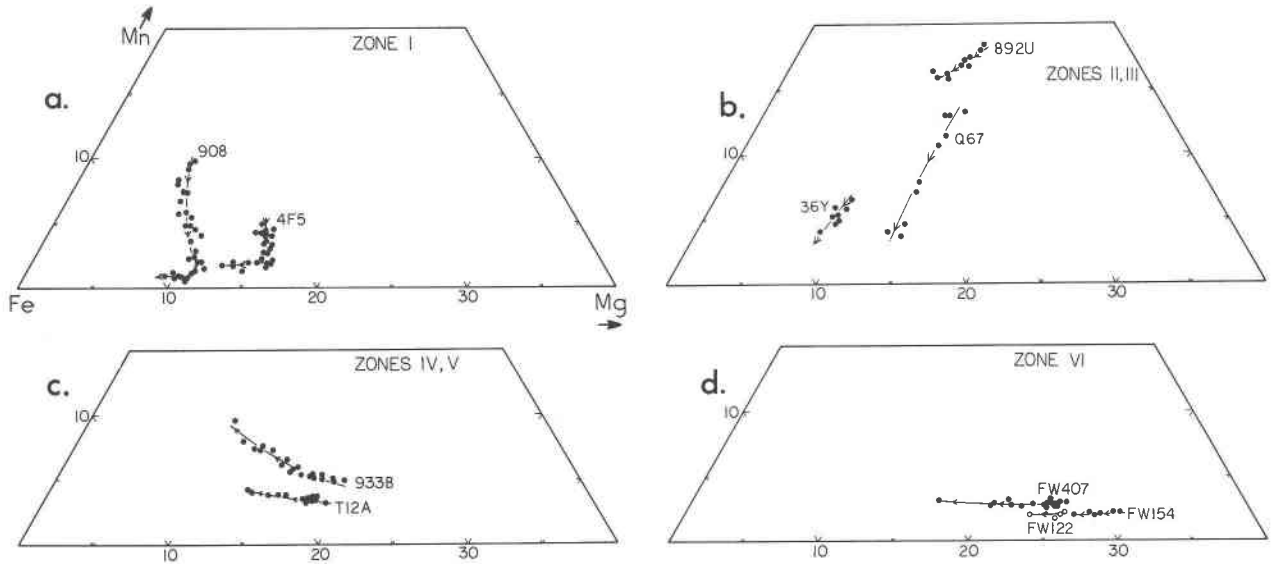


FIG. 3. Ternary plots of atomic percent Fe, Mg, and Mn in analyzed garnets. Arrows indicate direction of zonation from core to rim.



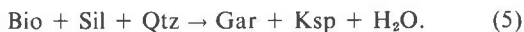
or ilmenite and rutile,



The limiting binary systems predict that $T_{\text{Fe}} > T_{\text{Mn}} > T_{\text{Mg}}$ for these reactions, which is consistent with the zoning in sample 892U (Fig. 3b).

The contours of composition for sample 933B in Zone IV (Fig. 4) show that the compositional variation is greatest in the marginal region of the garnet, in contrast to the more regular zoning across the lower-grade garnets. With regard to the distribution of Mn and Mg, the garnets from Zone IV may be considered to have reverse zoning from those in Zone I. The core to rim zoning trends for 933B and sample T12 from Zone V are shown in Figure 3c. Zoning from Mg-richer cores to Fe- and Mn-richer rims is strikingly different from zoning in inner parts of lower-grade garnets, though apparently similar to trends in outer parts of Zone I garnets. It is also similar to the garnet zoning observed by Grant and Weiblen (1971).

Garnet probably originated in Zones IV and V through operation of lower-grade continuous Fe-Mg-Mn reactions, as well as the continuous Fe-Mg-Mn dehydration reaction



For reaction (5), the predicted relative end-member temperatures are $T_{\text{Mg}} > T_{\text{Fe}} > T_{\text{Mn}}$. Consequently, the three-phase triangle Gar-Bio-Sil in the AKFM projection from KAlSi_3O_8 , SiO_2 , and H_2O (J. B.

Thompson, 1957) moves toward more Mg-rich compositions with increasing temperature. For Zone IV and V garnets, chemical zoning is strongest near the rims (Fig. 4), but is in the reverse sense from the trend predicted by this reaction. One explanation for this marginal zoning behavior, consistent with the shape of the garnets and zoning profiles (Fig. 3c), would involve late-stage operation of continuous reaction (5) in a retrograde sense:



For this case, reacting garnet rims are depleted in Mg relative to Fe and Mn.

The differences in element distribution in cores of garnets from Zones III, IV, and V compared to those from Zones I and II might be explained in a variety of ways. A first possibility is that the garnet in the higher-grade zones was not formed in these rocks at lower grade, either because of systematic bulk composition differences from the rocks now found in Zones I and II, or because they followed a different P - T path during metamorphism. The bulk composition argument can be ruled out, however, because bulk compositions in all zones, deduced from amounts of analyzed phases, overlap extensively. If the P - T path were the same and relict portions of lower-grade garnets remained within garnets of Zones III, IV, and V, then some internal mechanism, such as increased self-diffusion rates, could have caused elimination of the remnant internal-element distribution. The observation that zoning patterns

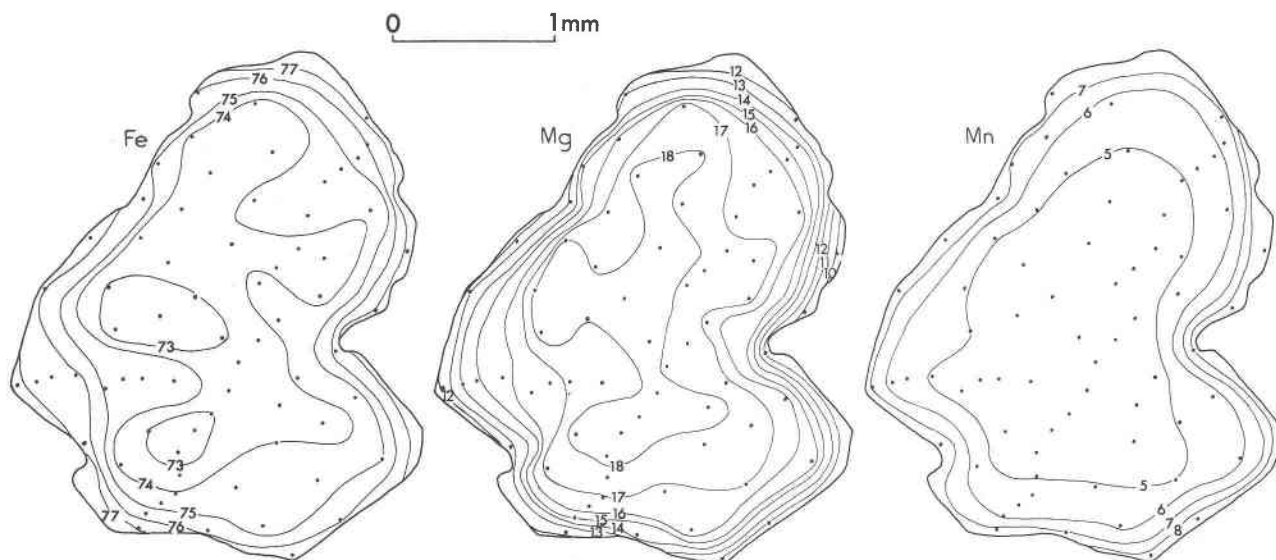


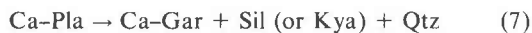
FIG. 4. Contours of atomic percent Fe, Mg, and Mn in a garnet from specimen 933B, Zone IV. Ca varies from 3.0 at edge to 4.4 in core. Dots indicate microprobe analysis spots.

are consistent with continuous reactions operating wholly within each zone may indicate that the higher-grade assemblages did not pass through the whole prograde sequence, but followed independent P - T trajectories.

Table 1 shows that Zones III, IV, and V contain the same AKFM assemblage (Gar-Bio-Sil) but are distinguished through the operation of continuous reactions involving Mus-Pla-Ksp-Sil-Qtz-H₂O (Evans and Guidotti, 1966; Tracy, 1975). While assemblages imply existence of the discontinuous reaction in a Ca-free system



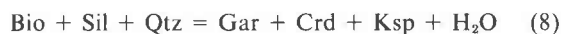
(A. B. Thompson, 1974a), the assemblage Mus-Pla-Ksp-Sil-Qtz is stabilized through continuous reactions involving Ca-exchange. In addition to continuous reactions due to Ca-Na-K exchange between Mus-Pla-Ksp, calcium is involved in a continuous reaction between plagioclase and garnet:



Data of Tracy (1975) suggest an equilibrium distribution of calcium between coexisting garnet and plagioclase in quartz-sillimanite bearing rocks, probably through operation of a continuous reaction such as (7) (see Appendix 2).

Zone VI

This zone is characterized by the occurrence of cordierite in pelitic rocks, possibly appearing by the discontinuous Fe-Mg-Mn reaction



In general, garnets from Zone VI are similar in appearance to those of Zones IV and V but are commonly larger, up to 3 cm or more in diameter. They are nearly homogeneous except where in contact with a more magnesian phase, biotite or cordierite, where a substantial decrease in Mg/Fe occurs. Similar observations have been made for garnets at equivalent grade in northern Connecticut and central Massachusetts by Hess (1971) and Richardson (1975). Zone IV, V, and VI garnets are significantly more pyrope-rich than those from lower grades, probably as a result of significant progress of reaction (5), one of two continuous Fe-Mg reactions which intersect to form the discontinuous reaction (8). Because T_{Fe} is only modestly lower than T_{Mg} for reaction (5), small temperature increases (or dehydrations) result in relatively large increases in pyrope content of garnet.

Figure 5 illustrates the compositional variation in a Zone VI garnet from sample FW407. In sample FW407, the biotite immediately adjacent to the garnet shows a slight increase in Mg/Fe. Richardson (1975) has suggested that this phenomenon may be due to a local cation-exchange process occurring in the first 100–200°C of cooling from the maximum metamorphic temperature. The Fe-Mg-Mn trends for these garnets (Fig. 3d) resemble those for Zone IV and V garnets and the near-rim trends for Zone I, and it is therefore tempting to ascribe all these effects to retrograde processes. Local retrograde cation exchange can only be demonstrated convincingly for

Zone VI garnets, while it is possible to explain the other trends by continuous reactions, either prograde or retrograde, involving participation of most of the phases in the rock with the exception of garnet cores. It is important, therefore, not only to document the compositional variations in garnets, but to attempt to relate the variations to the appropriate continuous or exchange equilibria.

Mineral composition and the estimation of temperature and pressure

Temperature estimates from $K_D(\text{Fe-Mg})$ data

Temperatures may be estimated using two-phase Fe-Mg cation thermometry if the distribution coefficient, $K_D(\text{Fe-Mg})$, shows a definite temperature dependence or, alternatively, from the compositions of phases involved in continuous reactions, if they can be calibrated. Values of $\ln K_D(\text{Fe-Mg})$ obtained from analyzed coexisting Gar-Bio and Gar-Crd pairs have been crudely calibrated as functions of T (A. B. Thompson, 1976b). The formulation necessarily assumes ideal solution of *all* components in the crystalline solution [$K_D(\text{Fe-Mg})$ is independent of composition] and that ΔC_p for the exchange reaction is zero.

Temperatures estimated from the $\ln K_D(\text{Fe-Mg})$ values obtained from microprobe analyses in this study, using the calibration of A. B. Thompson, are presented in Table 3. In contrast to the garnets, where core, intermediate, and rim compositions are available for K_D calculations, biotite was found to be essentially homogeneous within individual specimens, except in Zone VI where biotite immediately adjacent to garnet is more magnesian than elsewhere in the rock (Table 3), due to local retrograde exchange. It is reasonable to assume that K_D calculated from garnet-rim and matrix-biotite compositions gives an indication of the last continuous retrograde metamorphic equilibration. However, it is a serious problem to estimate how much the matrix-biotite composition might have been changed by retrograde (and prograde) reactions from the composition it had when it was in equilibrium with intermediate or core garnet compositions. It can be reasoned that the amount of biotite composition change during a retrograde continuous reaction would be proportional to the extent that bulk garnet participated in the reaction. In cases where the rock contains very little garnet and/or the retrograde outer rims are extremely narrow (and there are no other abundant Fe-Mg-Mn phases), the matrix biotite may have been close to

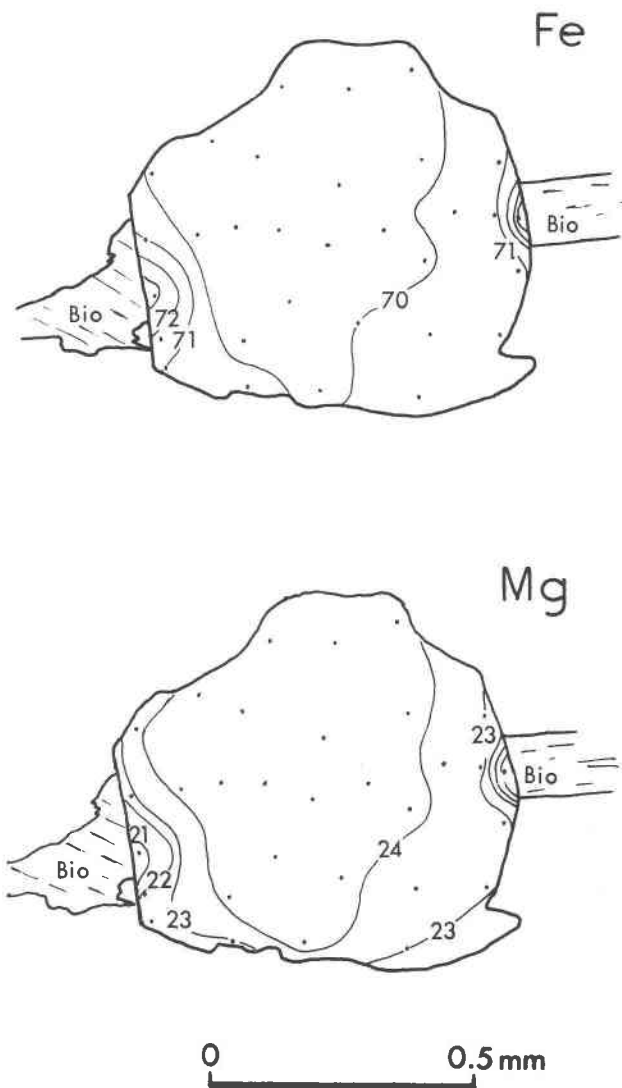


FIG. 5. Contours of atomic percent Fe and Mg in a garnet from specimen FW407, Zone VI. Ca and Mn are virtually homogeneous, 3.2–3.7 and 2.3–2.8 atomic percent respectively. Note the strong compositional change near adjacent magnesian biotite.

being an infinite reservoir, and there is no way that the biotite Fe/Mg ratio could have increased substantially from what it was earlier. Here, K_D determined from garnet interior and matrix biotite should give a substantially correct estimate of prograde temperature. On the other hand, when the rock contains very abundant garnet and/or shows very extensive retrograde rims as exemplified by specimen 933B (Table 2, Figs. 3c, 4), the Fe/Mg ratio of the matrix biotite should have increased substantially from what it was earlier. In this case, K_D calculated using interior-garnet and matrix-biotite compositions results in

TABLE 3. Mineral compositions and estimates of temperature and pressure

Zone	Spec.	Mineral	X_{Mg}	$\ln K_D$	T°C	Alm	Pyr	Spe	Gro	Pl _a X _{Ca}	Estimated P (kbar)			
											(a)	(b)	(c)	
I	908	Gar core	.076	2.13	470	78.7	6.5	9.0	5.9					
		Gar int.	.118	1.64	585	84.9	11.5	1.5	2.1			>5.6		
		Gar rim	.096	1.87	530	88.3	9.4	0.9	1.3					
		Bio	.410											
I	50Y	Gar core	.185	1.58	600	70.7	16.1	7.2	6.0	25	>6.2		6	
		Gar rim	.147	1.85	535	74.4	12.9	6.8	5.9					
		Bio	.524											
I	U94X	Gar core	.120	1.93	520	81.0	11.0	3.9	4.1					
		Gar int.	.150	1.67	580	79.4	14.0	3.3	3.9			>5.8		
		Gar rim	.103	2.06	490	83.4	9.6	2.8	4.1					
		Bio	.484											
I	4F5AY	Gar core	.146	1.71	570	75.8	13.0	4.6	6.7					
		Gar int.	.164	1.57	605	78.6	15.4	2.0	4.0					
		Gar rim	.125	1.89	525	81.9	11.7	2.0	4.4					
		Bio	.485									>6.1		
II	36Y	Gar core	.098	1.48	635	81.6	8.9	3.3	6.2		>4.9	<5.9		
		Gar rim	.087	1.61	595	85.0	8.1	3.8	3.0					
		Bio	.322											
II	Q67X	Gar core	.154	1.81	545	71.3	13.0	12.9	2.9					
		Gar int.	.144	1.90	525	77.8	13.1	3.3	3.8	23			5	
		Gar rim	.134	1.97	510	78.3	12.1	3.7	5.9					
		Bio	.527											
III	892U	Gar core	.148	1.66	580	66.9	11.6	17.6	3.9	27	>5.5	<7.2	4	
		Gar rim	.122	1.88	530	69.8	9.7	14.9	5.6					
		Bio	.477											
IV	871	Gar core	.168	1.40	660	70.4	14.2	11.6	3.8	33	>5.3	<7.6	3	
		Gar rim	.130	1.70	570	71.4	10.7	13.5	4.4					
		Bio	.449											
IV	869	Gar core	.159	1.47	640	68.7	13.0	13.8	4.6	31	>5.2	<7.4	5	
		Gar rim	.115	2.24	450	67.8	8.8	18.2	5.1					
		Bio	.450											
		Gar core	.214	1.41	655	64.3	17.5	12.8	5.4	40		>5.7	<8.5	4
IV	L11Y	Gar rim	.178	1.63	585	65.1	13.5	15.9	5.5					
		Bio	.526											
		Gar core	.188	1.34	680	73.2	17.1	6.7	3.0	23		>5.5	<8.0	4
		Gar rim	.180	1.40	660	73.6	16.3	7.1	3.0					
IV	933A*	Bio	.470											
		Gar core	.204	1.22	720	72.4	18.6	4.7	4.3	28		>5.6	<8.5	5
		Gar rim	.113	1.92	520	77.2	9.8	9.6	3.5					
		Bio	.465											
IV	595C	Gar core	.232	1.36	670	59.2	17.9	17.8	5.1	32	>5.4	<8.8	5	
		Bio	.533											
		Gar core	.175	1.39	660	76.5	16.2	3.7	3.6	16		>5.6		6
V	507B	Gar rim	.156	1.53	620	78.1	14.5	3.8	3.7					
		Bio	.460											
		Gar core	.194	1.42	650	70.6	17.1	9.4	2.9	21		>5.6		3
V	T10B	Gar rim	.149	1.74	560	74.0	12.9	10.2	2.9					
		Bio	.500											
		Gar core	.189	1.29	695	75.6	17.6	3.3	3.5	28		>5.6		4
V	T12A*	Gar rim	.139	1.66	580	79.9	12.9	4.1	3.1					
		Bio	.459											
		Gar core	.180	1.37	670	75.6	16.6	5.2	2.6	23		>5.6		2
		Gar rim	.110	1.94	515	80.2	9.9	7.4	2.5					
VI	FW283	Bio	.464											
		Gar core	.271	1.29	695	69.7	25.9	1.8	2.6	19		6.3		4
		Gar rim	.238	1.54	615	72.7	22.8	1.9	2.6					
VI	FW154	Bio	.595											
		Crd	.750	2.09	640									
		Gar core	.273	1.40	660	69.3	26.0	2.2	2.5			6.3		
		Gar rim	.236	1.60	595	72.9	22.4	2.1	2.6					
VI	FW122	Bio	.605											
		Crd	.720	1.92	695									
		Gar core	.258	1.32	685	69.6	24.2	2.8	3.4	27		6.2		5
		Gar rim	.243	1.40	660	70.9	22.8	2.9	3.4					
VI	FW407	Bio	.566											
		Crd	.710	1.95	685									
		Gar core	.261	1.33	680	69.7	24.7	2.2	3.4			6.3		
		Gar rim	.171	1.87	530	77.7	16.0	3.0	3.3					
		Bio rim	.600											

*Samples in which modal garnet is high relative to biotite (see text).

(a) Estimated from Gar-Crd-Al silicates-Qtz data (see text and Thompson, 1976).

(b) Estimated from Gar-Bio-Sil-Mus-Qtz data (Appendix 1).

(c) Estimated from Gar-Pl_a-Al silicates-Qtz data (Appendix 2).

a "false" high temperature estimate (Table 3, 933A, 933B, T12A).

Gar-Bio pairs from Zones I and II yield temperatures which are lowest for cores and outermost rims,

and highest for near-rim, maximum-Mg compositions. This is consistent with the previously stated model of interior zoning due to prograde continuous reactions and rim zoning due to retrograde continuous reactions. For Gar-Bio pairs from Zones III, IV, V, and VI, values of $\ln K_D(\text{Fe-Mg})$ using garnet interior compositions give temperatures 50 to 200°C higher than using rim compositions. The maximum temperatures appear to be about 580 to 605°C for Zones I and II, 600 to 675° for Zones III and IV, and 650 to 695° for Zones V and VI. As emphasized earlier, the absolute values of temperature cannot be estimated better than about $\pm 50^\circ\text{C}$ by these methods, although relative temperatures will be as indicated. These temperatures appear to be wholly consistent with those estimated from appropriate continuous reactions calibrated from experimental data (A. B. Thompson, 1976b).

Pressure estimates from continuous Fe-Mg reactions

The marked pressure dependence of some continuous reactions means that the compositions of coexisting phases may be used to predict pressure if the temperature can be independently evaluated, $a_{\text{H}_2\text{O}}$ is known, and the continuous reactions have been calibrated. One such continuous Fe-Mg-Mn reaction



can be described by the equation of state for one end-member reaction (either Fe, Mg, or Mn) and equations for two of the three binary exchange reactions, Fe-Mg, Fe-Mn, or Mn-Mg. The P - T curve for the pure-Fe end-member reaction (9) with sillimanite has been investigated by Richardson (1968, f_{O_2} buffered by QFM) and by Weisbrod (1973a, f_{O_2} buffered by QFI). The P - T curve for the pure-Mg end-member reaction has been calculated using a formulation for the pure-Fe data and the Fe-Mg exchange potential ($\Delta\mu^\circ_{\text{FeMg}}$) for Gar-Crd (A.B. Thompson, 1976b). The P - T curve for the pure-Mn end-member reaction has been investigated in synthesis experiments by Dasgupta *et al.* (1974). The location of this latter curve is in disagreement with the investigation of Weisbrod (1973b) on the Fe-Mn continuous reaction, but has been used in the present paper. P - X loops for the Fe-Mg, Fe-Mn and Mn-Mg binaries have been calculated using end-member molar volume data, experimental data on the P - T curves for the pure-Fe and pure-Mn reactions, and a calculated P - T curve for the Mg reaction, assuming ideal solution on equivalent sites (A. B. Thompson, 1976b, Fig. 2). These data are presented on a ternary Fe-Mg-Mn

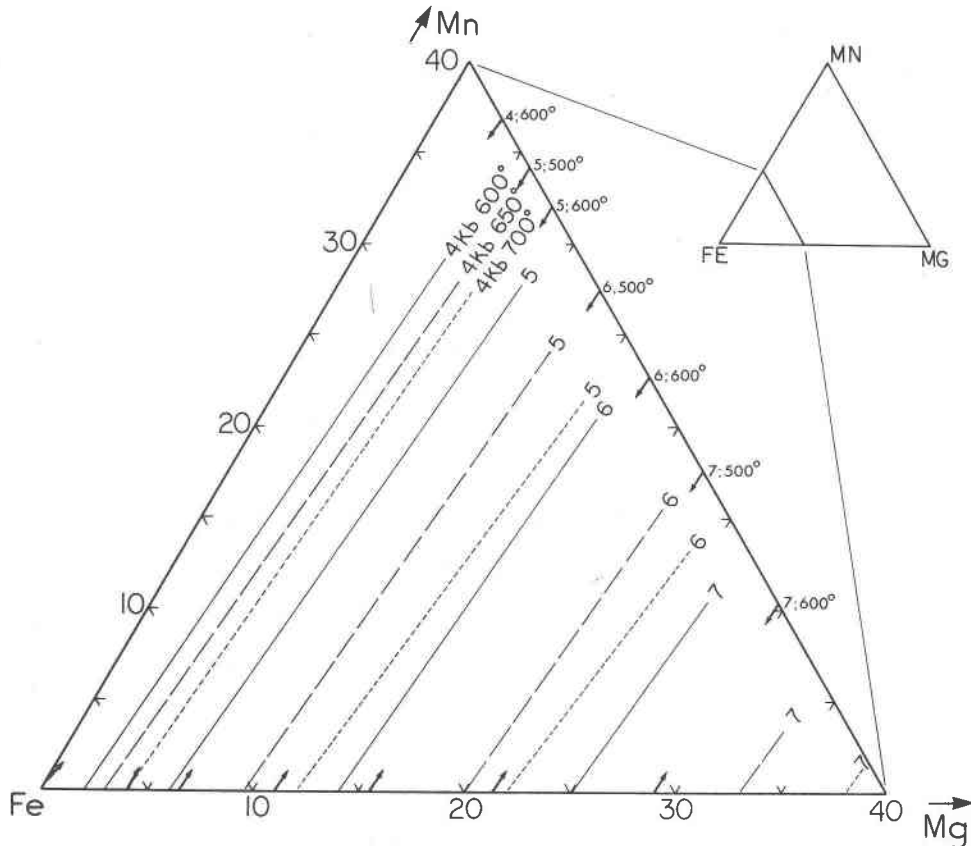


FIG. 6. Ternary Fe-Mg-Mn diagram with isopleths showing compositions (in atomic percent) of garnet in equilibrium with cordierite and sillimanite at given temperatures and pressures. See text for details on construction of this diagram. Arrows on edges indicate composition isopleths if Al-silicate is kyanite rather than sillimanite (4:600° indicates 4 kbar at 600°C).

diagram (Fig. 6) showing the temperatures and pressures at which garnets of the indicated composition would be in equilibrium with Fe-Mg-Mn Crd, Sil, and Qtz. Pressures determined from this Fe-Mg-Mn data necessarily give lower pressure estimates than if the experimental Fe-Mg data (corrected for Mn) of Hensen and Green (1973) or Currie (1971) had been used. The calculated Mg end-member reaction used here is about 3 kbar below that of Hensen and Green.

The assemblage Gar-Crd-Sil-Qtz-Ksp is known only in Zone VI, and yields estimates of pressure of 6.2-6.3 kbar (Table 3, column a). In the more common assemblage Gar-Bio-Sil-Qtz, the garnet is necessarily *more Fe-rich* than garnet in the assemblage Gar-Crd-Sil-Qtz at the same *P* and *T*. Consequently, due to the orientation of the Gar-Crd *P-X* loop, with garnet on the high-*P* side, Gar-Sil-Qtz (+Bio) *without* cordierite must indicate higher pressure than garnet of the same composition *with* cordierite. Garnet composition in the assemblage

Gar-Bio-Sil-Qtz, applied to Figure 6, should then yield an estimate of *minimum* pressure (Table 3, column a). Because reaction (9) is metastable in the kyanite zone, *P-X* loops and an Fe-Mg-Mn triangle were constructed for reaction (9) with kyanite (see caption Fig. 6), and pressure estimates for Zone I samples were obtained from these.

Other continuous reactions involving Gar-Bio-Sil-Qtz can be used for thermometry and barometry. If muscovite is present with Gar-Bio-Sil-Qtz, as in Zones II, III and IV, then continuous reaction (3), which has values of dP/dT for end-members approaching zero, could be a useful barometer, but not thermometer. This continuous Fe-Mg reaction has been calibrated (neglecting Mn because of the paucity of data) using the calculated dP/dT for the pure-Fe reaction, the location of an invariant point through which this curve must pass, and $\Delta\mu^\circ_{\text{FeMg-1}}$ to calculate the Mg reaction (see Appendix 1). The pressures estimated from this method

(Table 3, column b) are considered to represent maximum values because the effect of Mn would be to lower the pressure of the continuous Fe-Mg reaction. This calibration is even less dependable than Gar-Crd-Sil(Kya) because it is not tied to experimentally investigated equilibria.

A further attempt at quantitative geobarometry uses the compositions of coexisting garnet and plagioclase, with Al_2SiO_5 and quartz (see Appendix 2). Calibrations of reaction (7) have been used to obtain the pressure estimates shown in Table 3, column c.

Difficulties in use of zoned garnets for geothermometry and geobarometry

While the data presented here are considered to illustrate the importance of prograde continuous reactions in the formation of growth zoning in garnet, difficulties in interpretation are apparent. First, modelling of garnet growth zoning with such models as Rayleigh fractionation (Hollister, 1966) can be successful only if the garnet is involved in a single continuous reaction during its growth. Second, with available data, we are unable to discriminate fully between garnet zoning due to continuous prograde reactions, retrograde reactions either between adjacent phases or within a larger volume around the garnet, homogeneous diffusion within garnet, exchange with Fe-Mg-Mn inclusions, and reactions involving a possible melt phase at the highest grades.

The observed zoning of garnets from Zones IV, V, and VI cannot be successfully explained by prograde continuous reactions involving iron, magnesium, and manganese. However, as discussed by J. B. Thompson (1972, p. 32), simultaneous reactions involving iron-titanium oxides, iron sulfides, or graphite can displace projected bulk compositions in the AKFM tetrahedron away from or towards (FeO) and consequently affect the nature of the Fe-Mg continuous reactions. Similarly, as discussed by A. B. Thompson *et al.* (1976 and in preparation), simultaneous reactions involving calcium and sodium phases such as white micas, feldspars, epidote, and calcite can displace projected bulk compositions away from or towards Al_2O_3 and consequently displace the Fe-Mg continuous reactions. Reactions involving phases that display Tschermak's exchange, for example, $Al_2Mg_{-1}Si_{-1}$ in muscovite, biotite, chlorite, and possibly staurolite, will have the same effect. Such reactions operating simultaneously with Fe-Mg-Mn exchange reactions could actually lead to garnet consumption or a net change in zoning patterns con-

trary to those implied by continuous Fe-Mg-Mn reactions.

The role of diffusion in producing the observed zoning is difficult to assess. In fact, we have taken definite advantage of the slow diffusion rates within garnet that have led to the preservation of growth zoning, at least for Zones I, II, and III. This suggests that the exchange reactions within these samples have involved all the phases in the rock outside the garnet. This implied mobility of exchangeable species may be related to the presence of some transporting medium, such as the unseen "intergranular film." Further evidence that garnets from the lower-grade zones preserve continuous reaction zoning is the progressive decrease from core to rim of the ratio Mn/Fe in both host garnet and included ilmenite (see Fig. 2 and Table 4). For these coexisting phases, the ratio $(Mn/Fe)^{gar}/(Mn/Fe)^{ilm}$ changes slightly from core to rim. If this K_D could be calibrated as a function of temperature, then it might be possible to distinguish prograde continuous Fe-Mg-Mn reactions from retrograde two-phase cation exchange between garnet and ilmenite.

If mass balance across mineral contacts in exchange equilibrium can be evaluated, then the scale of equilibration can be determined. Moreover, the sense of the Fe-Mg exchange may be related to the change in the two-phase $K_D(Fe-Mg)$ and the appropriate relative temperature change deduced. The demonstration of total mass balance across garnet-cordierite and garnet-biotite junctions in Zone VI rocks (Richardson, 1975) and the absence of hydrated minerals at grain boundaries (Hess, 1971) is taken to indicate diffusional cation exchange between adjacent mineral grains only. The observed change in $K_D(Fe-Mg)$ indicates diffusion in response to *falling* temperature during a "dry" period of retrograde

TABLE 4. Compositions of ilmenite inclusions and adjacent garnet in specimen 908

Code*	Distance from center		Atomic percent			Mn/Fe	$\frac{(Mn/Fe)_{Gar}}{(Mn/Fe)_{Ilm}}$
			Fe	Mn	Mg		
A	0	Gar	.834	.097	.069	.116	7.2
		Ilm	.980	.016	.004	.016	
B	0.8 mm	Gar	.855	.060	.085	.070	7.0
		Ilm	.985	.010	.005	.010	
C	1.3 mm	Gar	.868	.034	.098	.039	7.8
		Ilm	.983	.005	.012	.005	
D	1.8 mm	Gar	.890	.008	.102	.009	9.0
		Ilm	.997	.001	.002	.001	

* See Figure 2a.

metamorphism (Lasaga and Richardson, 1975). Homogeneous interior compositions of Zone VI garnets could be taken to indicate that, during prograde growth, diffusion rates within the garnet exceeded the rate of growth on the exterior, or that increased homogeneous diffusion rates at the inferred temperatures (650–695°C) were able to eliminate earlier growth zoning.

If the near-rim zoning in garnets from Zone VI represents a retrograde exchange in response to cooling, it could be suggested that the near-rim zoning in garnets from Zone I and the core to rim zoning in garnets from Zones IV and V are also due to retrograde processes. The more concentric patterns of near-rim zoning in garnets of Zones I–V, however, which occurs irrespective of the adjacent phase, indicates a more complex mechanism than local two-phase cation exchange. Moreover, other phases in the assemblage appear to be internally homogeneous in composition. The reequilibration may have involved other phases in the rock, thereby representing retrograde continuous reaction, operating in a reverse sense from the prograde counterpart.

Speculations on significance of temperature and pressure estimates

Although the P and T estimates of this study are tentative and will, of course, be subject to major correction on the basis of additional and improved experimental work, it is useful to consider some of the geological implications. The estimates of temperature and of pressure (or minimum pressure) are plotted directly on a P - T diagram (Fig. 7) together with three conflicting sets of experimental data on the stability of the Al_2SiO_5 polymorphs (Newton, 1966; Richardson *et al.*, 1969; Holdaway, 1971). Minimum pressures for most zones lie within 1 kbar below estimated pressure for Zone VI and suggest that the last prograde equilibration may have been near 6 kbar over the entire area, well above any of the conflicting experimental triple points. Only specimens from Zone I have minimum pressures for estimated temperatures that lie on the kyanite side of *all* of the experimental Kya–Sil equilibria. All the Zone I specimens studied actually occur close to the kyanite–sillimanite isograd, and would be expected to fall close to this equilibrium in Figure 7.

Relict kyanite is fairly common in some rocks of Zone II, particularly near the Keene gneiss dome, in the north center of Figure 1. Detailed work on gedrite–cordierite gneisses in this vicinity (Robinson and

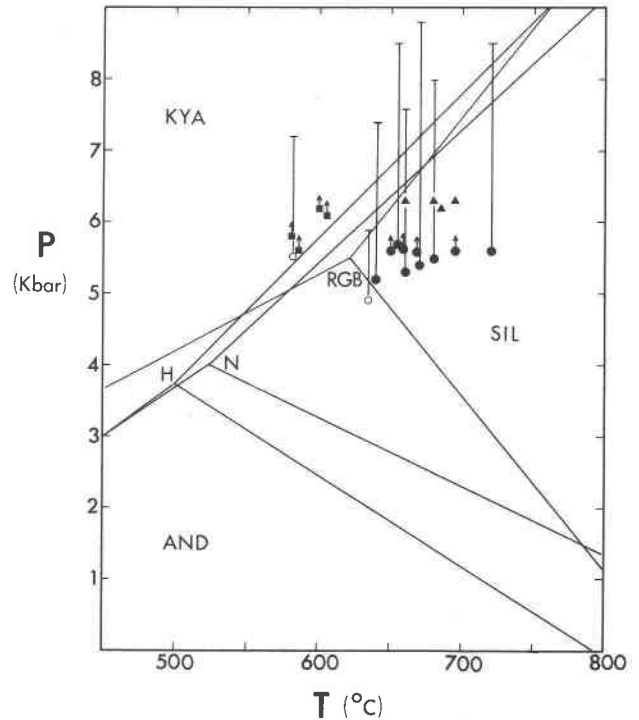


FIG. 7. P - T diagram summarizing the pressure and temperature estimates made for samples in this study: Zone I (squares), Zones II and III (open circles), Zones IV and V (closed circles) and Zone VI (triangles). Symbols with arrows are minimum pressure estimates. Bars are used for samples with minimum (from Gar–Crd–Sil[Kya]–Qtz) and maximum (from Gar–Bio–Mus–Sil–Qtz—see Appendix 1) pressure estimates. Al_2SiO_5 triple points shown for comparison are N (Newton, 1966), RGB (Richardson *et al.*, 1969) and H (Holdaway, 1971).

Jaffe, 1969) suggests late-stage crossing of the pressure-sensitive reactions

Gedrite + Sillimanite + Quartz



and

Gedrite + Sillimanite



to lower pressure, interpreted as the product of tectonic unloading associated with the rise of the gneiss domes.

Several of the rocks of Zone VI, including FW-283 and FW-122 (Field, 1975), contain columnar aggregates of sillimanite crystals with textural features similar to those at Gap Mountain, New Hampshire (Rosenfeld, 1969). These may be interpreted as sillimanite pseudomorphs after andalusite. Interestingly, the estimated temperature and pressure for

specimen FW-122 (Table 3) lie close to the "iso-meke" determined using quartz inclusion piezothermometry by Adams *et al.* (1975, Fig. 10) for specimen $\gamma 612$ from Gap Mountain. However, they suggest temperatures between 500 and 600°C for their specimen. Regardless of precise temperatures, it appears that the rocks forming much of this part of the Merrimack synclinorium followed a trajectory on the high-temperature side of the Al_2SiO_5 triple point and, at least in the case of Zone VI in Massachusetts, suffered rather severe late-stage compression to pressures above any of the experimental triple points. The overall impression of late-stage unloading, due to uplift, in the Bronson Hill anticlinorium, and simultaneous late-stage compression due to downbuckling in the adjacent Merrimack synclinorium is consistent with the presently available picture of late-phase regional tectonics and gravity in central Massachusetts (J. B. Thompson *et al.*, 1968; Kick, 1975).

Summary

We have attempted to illustrate the importance of continuous reactions involving the whole assemblage in the formation of growth zoning within garnets of medium- to high-grade metamorphism, and to emphasize that adequate petrologic characterization of natural samples is mandatory. Appropriate continuous reactions controlling the compositions of co-existing minerals must be identified and scales of equilibration considered.

We are unable at this time to assess the total effect of the amount of diffusional mass transfer (Blackburn, 1968; Anderson and Buckley, 1973, 1974), which is a function of additional unknown variables, including relative time of annealing, initial magnitude of concentration gradients between garnet and matrix, and complex relations between chemical potentials and diffusive fluxes. It appears that the minor zoning of Zone VI garnets represents diffusional cation exchange in the absence of H_2O . If the marginal zoning of Zone IV and V garnets and the near-rim zoning reversal of Zone I garnets are due to retrograde continuous reactions in the presence of an aqueous fluid or film and not the result of complex prograde continuous reactions not represented by the AKFM pelite projection (with Mn), then a continuum of retrograde effects during cooling may be indicated, and these assemblages may be used to deduce some aspects of the cooling history. It is not yet known if this fluid requires an external source or reflects prograde dehydration fluids unable to escape

and hence available for partial rehydration during a change in local temperature conditions.

Any attempt to refine the methods outlined here requires further calibrations from precise experimental studies, particularly P - T location of all end-member reactions and binary or ternary continuous reactions, and additional data on the activity-composition relations of the crystalline solutions, particularly cordierite. Also important are information on the nature and rates of diffusion in major metamorphic minerals and the mechanisms of crystal growth and intergranular diffusion. If the techniques described in this paper can be applied with greater precision, it will provide a powerful tool for better understanding the development of metamorphic rocks.

Appendix 1. Calibration of a Gar-Bio-Sil-Mus-Qtz Fe-Mg geobarometer

An attempt has been made to calibrate crudely the divariant isothermal P - X (Fe-Mg) loops for the continuous Fe-Mg reaction, involving ideal garnet and biotite solutions,



for use as a geobarometer. A value for dP/dT of +8 bars deg^{-1} for the pure Fe-reaction was estimated (Thompson, 1976b, Table 2). The P - T location of the Fe and Mg end-member curves is constrained by Fe and Mg invariant points generated by the intersections of



and



in the end-member systems. This intersection is close to that between (9) and the reaction



in end-member systems in P - T space. Thus, the pure-Fe reaction (3) is assumed to pass through the points 500°C, 2.8 kbar and 700°C, 4.2 kbar, even though metastable. By analogy with experimentally investigated and calculated reactions in the Mg-system, reaction (3) is assumed to pass through 500°C, 17.4 kbar and 700°C, 19 kbar, even though metastable in the pure Mg-system. Alternatively, the Mg-reaction (3) can be calculated from the P - T location of the Fe-reaction (3) and the (FeMg_{-1}) exchange potential for Gar-Bio (Thompson, 1976b, Fig. 1). Isothermal P - X (Fe-Mg) sections were calculated from these P - T

locations of end-member reactions and their molar volume changes. The values presented in Table 3 (column b) represent pressures calculated by these procedures, by matching X_{Mg} of garnet core and biotite to the calculated P - X sections. Although crude, they are considered to give *maximum pressure* estimates, because the presence of Mn should displace these equilibria to lower pressures.

Appendix 2. Calibration of a garnet-plagioclase- Al_2SiO_5 -quartz geobarometer

The calculated breakdown reactions for plagioclase at high pressures (Thompson, 1974b, Fig. 8) were used as a base for the calculation of isothermal P - X sections for reaction (7). One-site ideal solution was assumed for plagioclase, and solution of grossular was assumed ideal through Ca_3 -(Fe, Mg, Mn)₃ substitution. The X_{Ca} values of coexisting garnet and plagioclase with sillimanite and quartz were used to estimate pressures for the temperatures shown in Table 3. The resulting pressures, shown in column c in Table 3, were necessarily determined from the asymptotic limb of an exponential curve and as such carry considerable uncertainty.

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