

## Fluid-assisted modification of garnet composition along rims, cracks, and mineral inclusion boundaries in samples of amphibolite facies schists

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### ABSTRACT

Compositional zoning patterns in garnet from schists in central Vermont and western Connecticut suggest that prograde garnet growth was followed by an episode of compositional modification during fluid-assisted metasomatism. Prograde garnet zoning patterns are modified, with characteristic depletion of  $X_{\text{Grs}}$  and corresponding increases in  $X_{\text{Alm}}$ ,  $X_{\text{Spss}}$ , and  $X_{\text{Prp}}$ , in a region 50–150  $\mu\text{m}$  thick near the rim of garnet and along cracks and mineral inclusion boundaries. These regions of garnet compositional modification have a clouded appearance in thin section because they contain abundant secondary inclusions of both minerals and fluid 1–2  $\mu\text{m}$  in size. Metasomatic reactions among the matrix mineral assemblages produced an assemblage more hydrous than the original prograde assemblage. The dissolution and concomitant reprecipitation of garnet in these samples occurred at lower amphibolite facies conditions.

### INTRODUCTION

Garnet compositions are commonly a basis for thermobarometric calculations, and thus modifications to prograde garnet zoning have been the subject of many previous studies. Volume diffusion can modify garnet compositions at temperatures greater than ca. 550–650 °C (Loomis et al., 1985; Florence and Spear, 1991). The prograde composition of garnet around primary mineral inclusions, trapped during garnet growth, can be modified by exchange reactions (Tracy, 1982) and by net transfer reactions (Whitney, 1991). Yardley (1977) suggested that modifications to prograde garnet zoning could be regionally mapped and used to define an isograd comparable to the breakdown of muscovite to form sillimanite and feldspar. Zones of abrupt change in prograde garnet zoning patterns, such as those described in the present study, can be formed by sequential episodes of garnet growth without volume diffusion (Rumble and Finnerty, 1974), by discontinuous reactions (Thompson et al., 1977), and by resorption with volume diffusion (de Bethune et al., 1975; Tracy, 1982; Karabinos, 1984).

This study focuses on modifications to prograde garnet zoning patterns in two locations, representing two different metamorphic episodes. These garnets abruptly change composition in a zone 50–150  $\mu\text{m}$  wide at their rims, around a variety of mineral inclusions, and along some cracks in the garnet interiors. The zones are decorated by small mineral and fluid inclusions 1–2  $\mu\text{m}$  in diameter. These zones are interpreted to have formed by metasomatic dissolution and reprecipitation of preexisting garnet.

### Sample locations

Rocks in this study were sampled from two locations in the Acadian zone of western New England (Fig. 1). Peak metamorphic minerals west of the Proterozoic massifs were produced mainly during Middle Ordovician (Taconian) metamorphism and during Early to Middle Devonian (Acadian) metamorphism east of the massifs (Sutter et al., 1985). Both metamorphisms produced Barrovian facies series mineral assemblages. The Acadian metamorphic front (Fig. 1) represents a zone of transition from rocks in which the mineral assemblages, textures, and structures are dominantly the result of Taconian metamorphism to those in which they are due to Acadian metamorphism (Hames et al., 1991). Pre-Silurian rocks east of the Acadian front, as at the locality described below, locally contain polymetamorphic mineral assemblages that reflect both events.

Rocks within the Sharon Quadrangle and vicinity, northwestern Connecticut (Fig. 1), were initially metamorphosed at middle amphibolite-facies conditions (Zen, 1981; Wang and Spear, 1991) during the Taconian orogeny and were again metamorphosed at varying grades during the Acadian orogeny (Hames et al., 1991). Sample WH141 is a metamorphosed shale, part of the Wallom-sac Formation, which in New York, Massachusetts, and Vermont contains Middle Ordovician faunas (Ratcliffe, 1974; Finney, 1986).

The structure and stratigraphy of the Stafford Quadrangle and vicinity in east-central Vermont (Fig. 1) were described by Doll (1943), White and Jahns (1950), How-

ard (1969), and Ralph (1982), and the metamorphism of the area was described by Menard (1991). Devonian rocks exposed in the quadrangle were deformed and metamorphosed during the Acadian Orogeny. The major structural feature in the Stafford Quadrangle is the Stafford Dome (Fig. 1), which exposes a sequence of rocks increasing inward from chlorite grade to kyanite grade. The sample described here is from a region that Ferry (1990) proposed to have experienced high synmetamorphic fluid flow.

## Methods

Mineral compositions for the sample from Connecticut were determined at the Virginia Polytechnic Institute and State University on a Cameca SX50 electron microprobe. Areal maps of Fe, Mg, Mn, and Ca concentration were based on X-ray intensity data accumulated from wavelength-dispersive spectrometers. Similarly, mineral compositions for samples from Vermont were determined using the Jeol 733 Superprobe at Rensselaer Polytechnic Institute. Analytical conditions for spot analyses of all samples were 15 kV and 15–20 nA for a maximum of 40 s per element analyzed. Natural and synthetic silicates and oxides were used as standards. FTIR and Raman spectroscopic study was conducted at Virginia Polytechnic Institute and State University.

## SAMPLE DESCRIPTIONS

### Taconian garnet overprinted by Acadian metamorphism, Sharon, Connecticut

Sample WH141 (Fig. 1; UTM coordinate B33234370) is a schist in which Taconian metamorphism produced the assemblage kyanite + staurolite + garnet + biotite + quartz + muscovite + plagioclase + ilmenite with minor apatite, graphite, and pyrite (Hames, 1990). Compositional layering in the exposure defines a crenulated, composite surface that is overgrown by the garnet, kyanite, and staurolite ( $T_3$  porphyroblasts of Sutter et al., 1985; Hames et al., 1991). This composite surface and the porphyroblasts are deformed by both northwest- and northeast-trending Acadian folds (the  $D_4$  and  $D_5$  folds of Ratcliffe and Harwood, 1975).

Porphyroblastic garnet, kyanite, and staurolite have irregular, rounded shapes and are surrounded by coarse, randomly oriented muscovite, suggesting resorption of early-formed, less hydrous porphyroblasts and formation of a more hydrous assemblage. Traces of acicular sillimanite are present on the resorbed margins of garnet porphyroblasts in sample WH141. Plagioclase porphyroblasts are texturally zoned, with fractured, inclusion-rich cores and clear subhedral rims; however, they have a uniform composition of about  $An_{35}$ . The porphyroblast resorption, coarse muscovite, and acicular sillimanite are interpreted as having formed during Acadian metamorphism (Hames et al., 1991). The rim compositions of porphyroblasts in WH141 define  $P$ - $T$  conditions of  $510 \pm 25$  °C and  $4 \pm 0.5$  kbar; from this sample locality, Acadian metamorphic  $P$ - $T$  conditions increased mono-

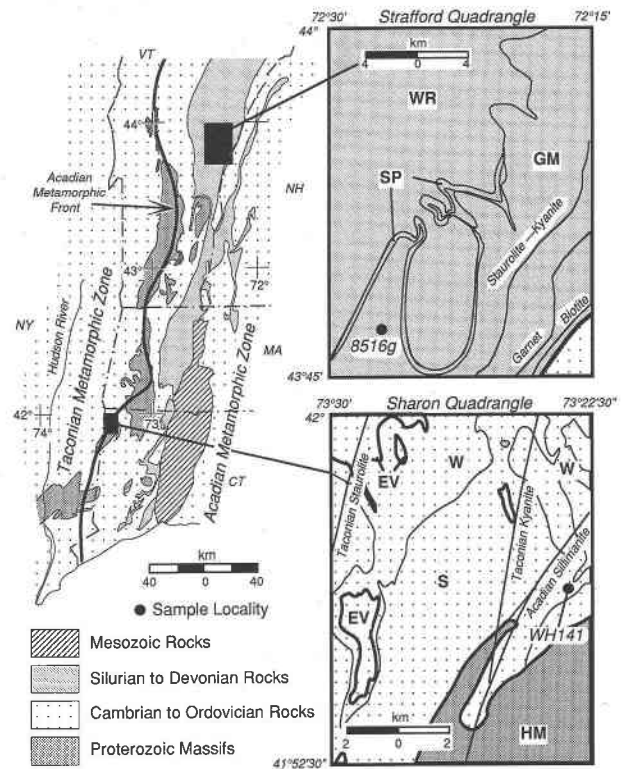


Fig. 1. Regional geologic maps of western New England indicating the general areas of Taconian and western Acadian metamorphisms (adapted from Sutter et al., 1985). One sample is from post-Taconian metasedimentary rocks of the Acadian zone in Vermont, and the other is from the Taconian-Acadian polymetamorphic zone in northwestern Connecticut. Geology of the 15' Stafford Quadrangle is based upon Doll (1943), White and Jahns (1950), Howard (1969), Rolph (1982), and Menard (1991): WR = Waits River Formation, SP = Standing Pond Volcanic Member, GM = Gile Mountain Formation. Geology of the 7½' Sharon Quadrangle is based upon Gates (1959), Zen (1981), and Hames et al. (1991): HM = Housatonic Massif, EV = Everett Formation, S = Stockbridge Formation, W = Wal-loomsac Formation.

tonically to greater than 650 °C and 7 kbar southeastward along the present erosion surface (Hames et al., 1991).

Textures of garnet representative of this locality are shown in Figure 2. The garnet interior is characterized by concentric changes in major cation concentrations that are consistent with growth zoning. In contrast, there is an abrupt decrease in  $X_{Grs}$  and accompanying increases in  $X_{Alm}$ ,  $X_{Spss}$ , and  $X_{Prp}$  within  $\sim 100$   $\mu$ m of the garnet rim. Identical compositional changes in garnet occur along some cracks and around some primary apatite, muscovite, biotite, and ilmenite inclusions. These compositional changes are uniform and do not vary with respect to which mineral is in contact with garnet along the rim or inclusion interface.

The regions of sharp compositional change are marked by abundant secondary fluid and mineral inclusions, up to  $\sim 2$   $\mu$ m in diameter, that impart a clouded appearance

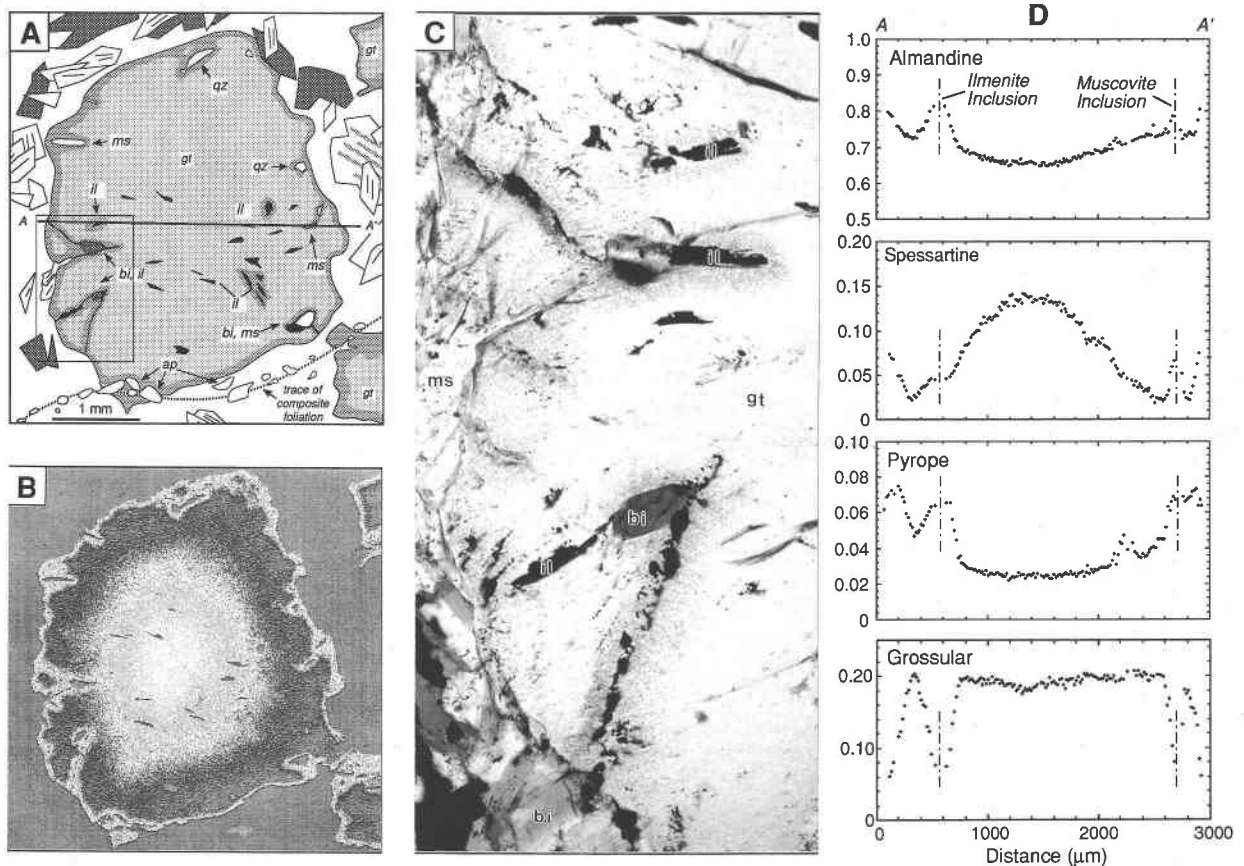


Fig. 2. Garnet in the Taconian-Acadian zone in northwestern Connecticut. (A) Sketch of microtextural relationships in sample WH141. The shaded zone around the garnet surfaces indicates the distribution of micrometer-scale fluid and mineral inclusions discussed in text. The box indicates the area of the photomicrograph in C. Symbols: ms = muscovite, ap = apatite, qz = quartz, il = ilmenite, gt = garnet, A–A' = traverse in D. (B) Mn X-ray map of the garnet porphyroblast in A (A and B are at the same scale). The brighter shades in B indicate higher Mn-content. Note that the increase of Mn near the garnet surface corresponds to the zone of micrometer-scale mineral and fluid

inclusions in A and C. The overall zoning patterns for  $X_{\text{Alm}}$ ,  $X_{\text{Prp}}$ , and  $X_{\text{Grs}}$  in this porphyroblast are the same as shown for  $X_{\text{Sp}}$  in B. (C) Photomicrograph of part of the garnet in A and B; note the correspondence between inclusion zones, cracks, and inclusion boundaries between B and C. (D) Microprobe compositional traverse across the garnet (vertical axes are in mole fractions). Note that each garnet component changes abundance in the shaded zones outlined in A–C; the vector of change for each garnet component is uniform within the garnet boundary zones that show Mn enrichment in B.

in thin section (Fig. 2A, 2C). The clouded regions along cracks in the garnet cross the concentric growth zoning pattern, and thus these small inclusions are secondary with respect to prograde garnet growth. Mineral inclusions in these regions are highly birefringent and appear to be muscovite and sphene. The fluid inclusions are generally equant, and negative crystal shapes (reflecting the host isometric structure) are common. Spacing between these inclusions ranges from 2 to 10  $\mu\text{m}$ . Some inclusions are arranged in parallel, linear arrays: these arrays are generally perpendicular to the garnet surface. The largest fluid inclusions ( $\sim 1\text{--}2 \mu\text{m}$  in diameter) have distinct vapor bubbles that adhere to the inclusion wall, suggesting that the fluids are  $\text{H}_2\text{O}$ -rich. FTIR and laser Raman spectroscopic data from garnets in sample WH141 (and oth-

ers in the vicinity) detect both  $\text{CO}_2$  and  $\text{H}_2\text{O}$  in the fluid inclusions.

#### Garnet in the prograde Acadian zone, Sharon, Vermont

Sample 8516g is a calcic pelitic schist from the Devonian Gile Mountain Formation collected from a roadcut on the northbound entrance ramp of I-89 in Sharon, Vermont (Fig. 1). The matrix assemblage is kyanite + staurolite + garnet + biotite + plagioclase ( $\text{An}_{23}$ ) + quartz + muscovite + graphite, with minor rutile + ilmenite + tourmaline + zircon. Ilmenite and the first generation of muscovite and biotite define a crenulated  $S_1$  schistosity. Orientation of a second generation of biotite defines the  $S_2$  schistosity. The garnet core has straight inclusion trails,

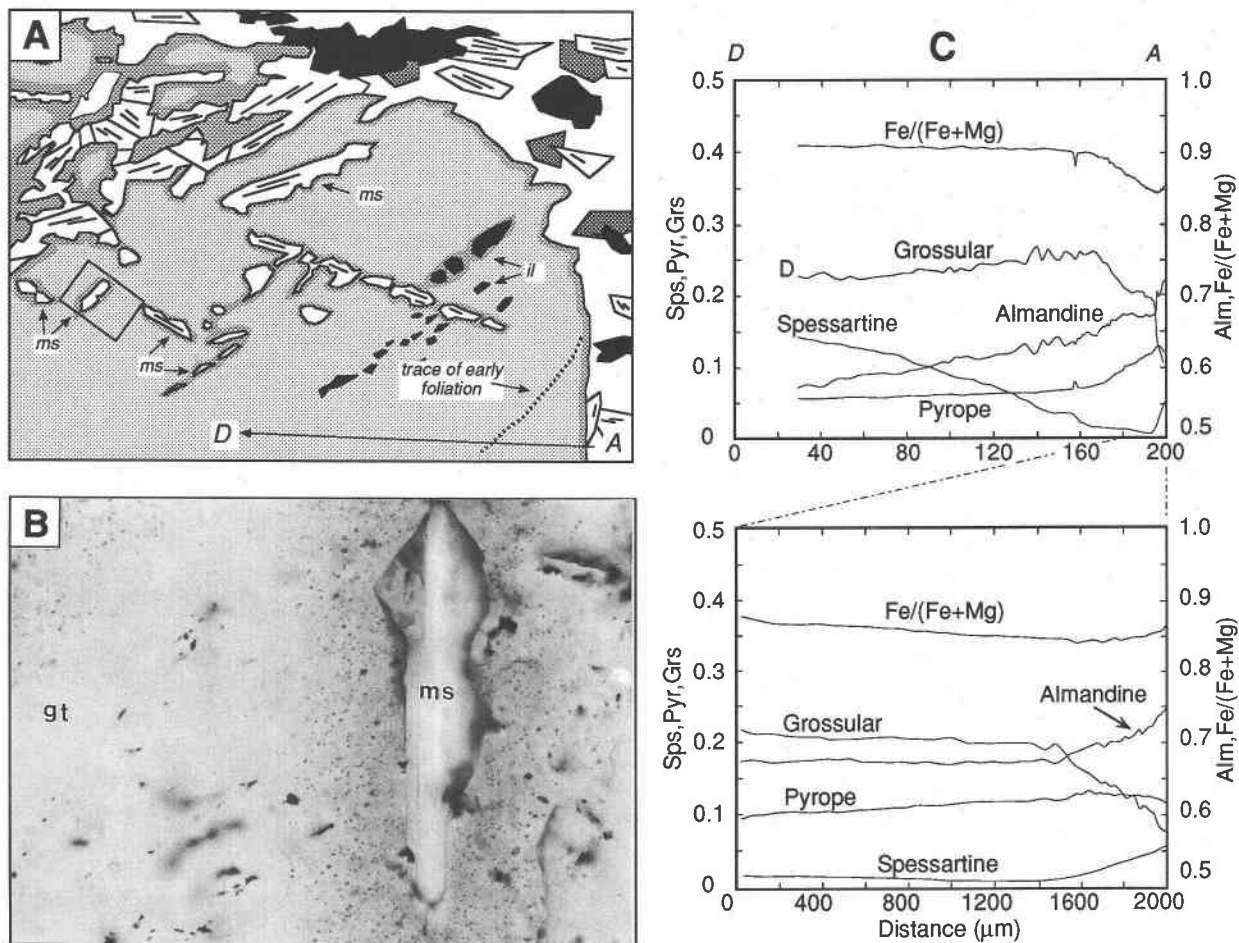


Fig. 3. Garnet from the Acadian zone in central Vermont. (A) Sketch of garnet in 8516g. Late, coarse muscovite porphyroblasts, appearing as inclusions in the section, crosscut the garnet and the included  $S_1$  surface (defined principally by ilmenite inclusions). The distribution of micrometer-scale fluid and mineral inclusions is indicated by the darker shaded pattern as in Fig. 2A. Line A–D refers to the compositional half-profile in C; the inset represents the location of the photomicrograph in B.

(B) Photomicrograph of a muscovite inclusion and surrounding garnet showing the distribution and scale of the fluid and mineral inclusions. (C) Garnet compositions in sample 8516g (profile lines are interpolated between 116 data points). Note the sharp decrease of  $X_{Grs}$  and increase of  $X_{Alm}$  and  $X_{Sps}$  from D to the garnet rim at A. Identical compositional changes occur around mineral inclusions, as shown in B. The 116 analysis points are along the traverse.

composed of epidote and graphite, that define an included  $S_1$  surface at a high angle to the matrix  $S_2$  foliation (Fig. 3).

Garnet in sample 8516g is intensely resorbed (Fig. 3) and replaced along its rim and interior by coarse, unoriented muscovite that crosscuts the internal  $S_1$  fabric in the garnet (Fig. 3). The garnet rim and surfaces of the garnet adjacent to the late muscovite have a zone 30  $\mu\text{m}$  wide with abundant micrometer-scale fluid and mineral inclusions. Garnet compositions (Fig. 3C) are similar to those in sample WH141.  $X_{Grs}$  in the garnet core is high and fairly uniform. However, in the outer 50  $\mu\text{m}$  of the garnet rim,  $X_{Grs}$  decreases sharply as  $X_{Alm}$  and  $X_{Sps}$  increase; similar sharp zoning patterns are evident adjacent to some muscovite, ilmenite, and plagioclase inclusions.

Garnet in this sample grew during Acadian metamorphism by the reaction (Menard, 1991)  $\text{Chl} + \text{Ep} + \text{Ms} + \text{Qtz} + \text{Gr} = \text{Grt} + \text{Bt} + \text{Pl} + \text{H}_2\text{O} + \text{CO}_2$ . Garnet was interpreted to have grown along a computed  $P$ - $T$  path of heating during a 1-kbar pressure increase up to conditions of  $550 \pm 25$   $^\circ\text{C}$  and  $7.5 \pm 0.6$  kbar (Menard, 1991). The subsequent development of sharp compositional zoning at the rim of the garnet probably formed without a large change in  $P$  and  $T$ : the distribution coefficient of Ca between garnet and plagioclase inclusions does not change substantially across the sharp compositional zoning in the garnet. The large change of garnet and plagioclase compositions under seemingly isothermal, isobaric conditions indicates open-system behavior and metasomatic loss of Ca from the rock.

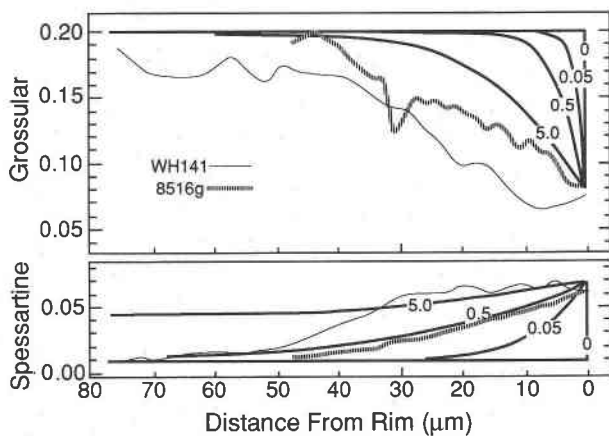


Fig. 4. Comparison of measured garnet zoning data with compositions predicted for volume diffusion (Florence and Spear, 1991) for various times at a constant temperature of 550 °C (thin lines). Compositions used in modeling are representative of the natural samples in this study. The initial model profile (0 m.y.) has flat zoning in the core and a step decrease in  $X_{Grs}$  at the rim. The measured  $X_{Sps}$  profile for sample 8516g fits the predicted curves at 0.5 m.y., but the poor fit for  $X_{Grs}$  for both samples at all model times suggests that the modification of  $X_{Grs}$  has been enhanced beyond that reasonable for simple volume diffusion.

#### PROCESSES OF GARNET RESORPTION

In addition to textures showing early garnet growth followed by later garnet resorption and replacement by a more hydrous assemblage, two features must be accounted for: (1) abrupt compositional zoning in a region 50–150  $\mu\text{m}$  wide near the garnet rims, cracks, and a variety of mineral inclusions; and (2) abundant mineral and fluid inclusions within these regions 0.5–2  $\mu\text{m}$  in size. Interpretations for the processes that formed these features fall within two broadly different mechanistic groups: intracrystalline volume diffusion and microscale metasomatic replacement. Our preferred interpretation of the textures is that the zones of inclusions formed concomitantly with the accompanying compositional changes by microscale infiltration metasomatism. Modification of garnet composition along external rims and cracks and locally around apatite, muscovite, ilmenite, and plagioclase inclusions (cf. Figs. 2, 4) was controlled by access of this fluid.

#### Compositional modification by volume diffusion

One interpretation for the observed sharp compositional zoning of the garnet near its rim is that it results from volume diffusion within existing garnet following a change of equilibrium compositions. This phenomenon has been documented in middle amphibolite and higher grade settings where temperatures exceeded ca. 650 °C. Several studies have concluded that temperatures below ca. 600 °C are insufficient to produce significant modification of prograde garnet zoning in garnets larger than a few millimeters in diameter for the duration of most metamorphic episodes (e.g., Florence and Spear, 1991). Thermobarometry from each of the present sample set-

tings indicates that temperatures did not exceed 550 °C during the garnet resorption episodes, and thus it is difficult to account for the observed changes by volume diffusion. Previous studies have also suggested that surface diffusion, assisted by an intergranular metamorphic fluid, may have enhanced compositional changes along garnet rims (Karabinos, 1984).

Models of multicomponent volume diffusion generally predict a smooth, curved compositional zoning profile as a result of modifications to growth zoning patterns (Florence and Spear, 1991, their Fig. 5). Ordinary volume diffusion tends to smooth out sharp shoulders and compositional breaks in zoning patterns. In contrast, the  $X_{Grs}$  profile from the samples of this study have a straight-line decrease of  $X_{Grs}$  at the rim that is separated from the core compositions by a shoulder—a distinct change in the slope of the zoning profile. Ordinary volume diffusion would tend to smooth out such a shoulder. The natural zoning profiles also contrast with typical patterns of volume diffusion in that the magnitudes of modification of  $X_{Sps}$  and  $X_{Grs}$  in these samples are quite similar. In contrast, the magnitude of modification for  $X_{Sps}$  predicted for volume diffusion is much greater than for  $X_{Grs}$  at any specific time. Relative to the modification of  $X_{Sps}$ , the observed modification of  $X_{Grs}$  was enhanced over that expected for simple volume diffusion. Thus the petrologic and textural data suggest that diffusion mechanisms alone are not responsible for the modifications to growth zoning in the present study samples.

#### Fluid-assisted metasomatism of garnet

The prominent features discussed earlier indicate a mineral replacement process across a reaction layer 50–150  $\mu\text{m}$  wide in which a metamorphic fluid participated—a metasomatic process. Lindgren (1928) defined metasomatism as a process of mineral replacement by simultaneous dissolution and reprecipitation, with accompanying mass transfer of components, via capillary metamorphic fluids. A dissolution reaction can occur most readily upon mineral surfaces where intracrystalline defects (especially vacancies) are concentrated. Previous studies have documented preferential dissolution of a mineral in pits or tubes that formed on surface defects (e.g., Blum et al., 1990, and references therein). Mineral growth can be promoted along the edges of pits, even as the garnet is being resorbed, because of their high surficial energy and resulting thermodynamic instability. Local equilibrium during metasomatism (Thompson, 1959; Korzhinskii, 1970) could fractionate cations between the garnet edges, other minerals, and the metamorphic fluid as the garnet dissolves and reprecipitates. If the equilibrium was controlled by the fluid, then the equilibrium composition of the reprecipitated garnet should be the same along every surface to which the fluid had access, irrespective of which minerals are in contact with the garnet. This metamorphic fluid could also be trapped during the metasomatism as pits are sealed by growth along their edges.



We propose that this single-crystal style of metasomatism formed the textural and compositional discontinuities along garnet surfaces in these samples. The garnet resorption textures, presence of fluid inclusions, and accompanying compositional changes suggest that the outer surface of the garnet was dissolved in a metamorphic fluid. The garnet being resorbed probably had relict, disequilibrium compositions and the equilibrium garnet compositions changed as a result of the resorption. This mineral-surface scale metasomatism modified the original prograde garnet zoning pattern along every pathway accessed by the metamorphic fluid, even along cracks and some inclusion boundaries inside the garnet. Reprecipitation of garnet in equilibrium with the fluid, concomitant with the dissolution, trapped both fluid and mineral inclusions in pits near the garnet surface.

We cannot prove that the new garnet was in equilibrium with the fluid, yet fluid-controlled equilibrium is strongly suggested because (1) garnet composition is similar along every surface that contains the mineral + fluid zone, irrespective of which mineral is in contact with garnet across the zone; (2) we obtain consistent pressure-temperature estimates (i.e., consistent mineral  $K_{eq}$  and thus  $P$ - $T$  estimates) from the garnet rims and matrix minerals in these samples; and (3) the  $P$ - $T$  estimates based upon garnet rim and matrix mineral compositions are compatible with the sample matrix assemblages and the regional metamorphic setting.

### IMPLICATIONS

These samples provide evidence of compositional modification interpreted to result from fluid-assisted metasomatism in metapelitic rocks at lower to middle amphibolite facies. The extent to which processes more rapid than volume diffusion can modify composition in minerals places limits on the applicability of models based on volume diffusion. Thus the validity of a volume diffusion model to establish guidelines for which natural minerals are safe to analyze in polymetamorphic rocks (Jiang and Lasaga, 1990) relies upon the extent to which volume diffusion is the fastest process for effecting a change in a particular garnet sample. Tracy (1982) suggested that hydration reactions can modify garnet rim compositions more effectively than volume diffusion. This study documents that these reactions occurred at amphibolite facies conditions and were metasomatic in the sense of Lindgren (1928). This study also emphasizes and expands a conclusion of Loomis (1983) and Whitney (1991): postentrapment net-transfer reactions can occur among garnet, primary mineral inclusions, and the matrix if there are cracks or inclusion boundaries that provide a link with the matrix assemblage.

The data of this study are from two areas in the New England Appalachians. In addition, previous studies in a variety of metamorphic settings have documented zones 10–300  $\mu\text{m}$  wide enriched in fluid and mineral inclusions that precede a reaction front. Zhou and Fyfe (1989) noted that an inclusion zone precedes reaction fronts in altered

volcanic glass. Features analogous to the present study have been reported in a separate part of Vermont by Karabinos (1984), in the Italian and Swiss Alps by de Bethune et al. (1975), and in the Dalradian of Scotland by Yardley (1977), indicating that this style of garnet compositional modification is more extensive than previously recognized.

### CONCLUSIONS

Garnets from a prograde metamorphic zone in Vermont and a polymetamorphic zone in Connecticut changed composition by a similar, crystal-scale metasomatic process. This metasomatism dissolved garnet with disequilibrium compositions and reprecipitated garnet with a composition in equilibrium with the metamorphic fluid and the evolving matrix assemblage. Fluid and mineral inclusions trapped within garnet and associated abrupt compositional changes are the principal indications of this process. The metasomatism also changed the composition of garnet along cracks that linked the garnet interior with the matrix.

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