

CONTACT METAMORPHISM OF CALCRETE IN NEW HAVEN ARKOSE (Tr/J): ROLE OF INFILTRATION METASOMATISM AND HYDROTHERMAL BOILING

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INTRODUCTION

The Upper Triassic/Lower Jurassic New Haven Arkose is a continental red bed deposit that locally contains abundant paleosol calcrete horizons. The calcrete occurs mainly as irregular nodules, lenses and root casts. At a quarry in Mount Carmel, Connecticut, calcrete-bearing arkose is exposed adjacent to a diabase dike. The dike extends upward from the Mount Carmel Sill, which is considered part of the larger West Rock Sill.

Emplacement of the dike and sill at ~200 Ma caused contact metamorphism of the sediments. Circulation of heated sedimentary pore fluids caused severe metasomatism of the pedogenic calcrete. Reactions among the carbonate, the surrounding siliciclastic sediments, and metasomatic fluids caused growth of calc-silicate minerals, such as epidote and grossular-andradite (grandite) garnet, producing a skarn-like mineral assemblage. Hydrothermal alteration minerals such as chlorite, clay minerals and copper sulfides were also formed.

Petrographic observations reveal evidence of intense infiltration metasomatism, such as fractures filled with calc-silicate minerals, and broken crystals of garnet and epidote. Electron microprobe analyses show that many garnet crystals display oscillatory compositional zoning, another indication of infiltration metasomatism. The compositional oscillations may indicate cyclical changes in a property or properties of the metasomatic fluid. Such changes could have been produced by hydrothermal boiling events in which overpressured pore fluids caused hydrofracturing of the partly lithified sediments and calcrete (or of early formed calc-silicates lining fluid-filled voids). This in turn would have caused a sudden drop in confining pressure, allowing the fluid phase to boil and release accumulated volatiles. In addition to the drop in pressure, such events could have caused significant changes in other fluid properties, such as f_{O_2} , pH and temperature.

GEOLOGIC SETTING

The rocks which are the subject of this study are located in the Hartford Basin of central Connecticut (see figure 1). The Hartford Basin is one of a series of Mesozoic basins found along the east coast of North America that are thought to have been formed by rifting of the supercontinent Pangaea in the early stages of the opening of the Atlantic Ocean. Structurally, the Hartford Basin is a half graben. It is bounded on the east by a west-dipping normal fault that separates the Triassic/Jurassic rocks of the basin from Paleozoic schists and gneisses of the New England Upland (see figure 2). The western boundary of the basin is an erosional contact, with Mesozoic sediments lying unconformably on Paleozoic basement rocks.

The Hartford Basin contains a series of sedimentary and volcanic rocks that belong stratigraphically to the Newark Supergroup (Froelich and Olsen, 1985). The New Haven Arkose is the bottom-most unit in the basin. It consists of tan to reddish sandstones, siltstones and conglomerates that are interpreted as fluvial, braided stream deposits and floodplain deposits (Hubert, 1978). The New Haven Arkose locally contains pedogenic calcrete horizons. Hubert (1977, 1978) found that paleosol calcretes are widely distributed in the arkose. Other rocks in the basin include diabase dikes and sills, basalt flows, and lacustrine grey sandstones and siltstones and black shales. The New Haven Arkose is estimated to be about 2200 meters thick; the total thickness of the rocks in the basin is thought to be at least 4 kilometers.

GEOLOGY OF SAMPLE LOCATION

The site which is the subject of this study is an inactive trap rock quarry located in the village of Mount Carmel, Connecticut (see inset in fig. 1). A private sanitation company, Dee's Inc., currently occupies the site. The rocks exposed in the quarry are baked sandstones and siltstones belonging to the lower member of the New

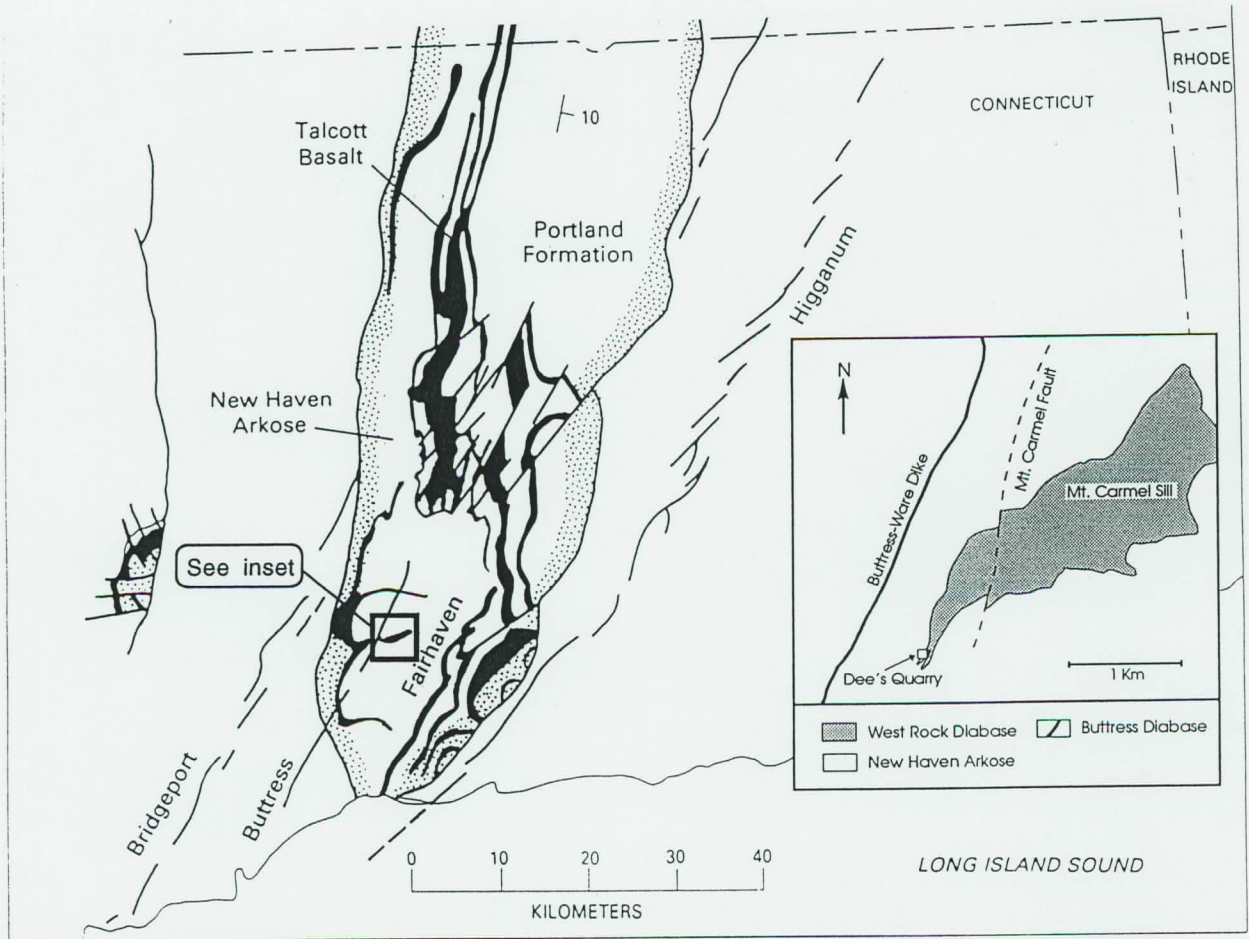


Figure 1. Map of Hartford Basin (after Philpotts, 1985). Inset shows sample location.

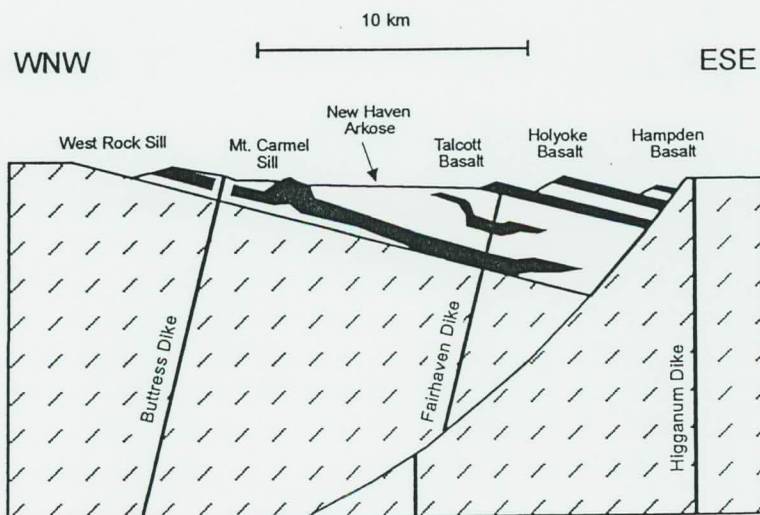


Figure 2. Cross section of Hartford Basin (after Philpotts and Martello, 1986).

Haven Arkose, as mapped by Fritts (1962), and a dike of West Rock Diabase. The dike is about 20–25 meters thick, strikes approximately N 30°E, and dips about 50° to the NW. The dike extends SW from the Mount Carmel intrusion. A small outcrop of arkose is exposed on the north side of the dike; south of the dike a larger expanse of arkose is exposed, with bedding striking about N 10°E and dipping gently (about 15°) to the east.

Irregularly shaped greenish patches containing epidote, calcite, and sometimes grandite garnet are present throughout the arkose at the site. These calc-silicate patches are interpreted as contact-metamorphosed calcrite nodules. The sizes, shapes and distribution of the altered nodules in the arkose are similar in appearance to unmetamorphosed calcrites exposed elsewhere in the Hartford Basin. In particular, epidotized root casts found at the quarry constitute convincing evidence that the sediments there are paleosols that contained pedogenic calcrite.

PETROGRAPHY

There are three main types of contact-metamorphosed calcrite (*meta-calcrite*) in the arkose at Dee's quarry, based on mineral assemblages. These types are: (1) epidote meta-calcrite, (2) garnet meta-calcrite, and (3) garnet-epidote meta-calcrite. The petrography of the three meta-calcrite types is described below.

Epidote Meta-Calcrite. The epidote meta-calcrite occurs in rounded and irregular nodules, lenses and layers. The nodules range in size from ~3 mm to about 5 cm; lenses and layers may be up to about 4 cm in thickness. The minerals that make up the meta-calcrite are epidote, calcite, hematite, chlorite, an authigenic clay mineral, and albitic plagioclase. Hematite occurs both as relatively coarse silvery-black plates (specular hematite) and as tiny reddish or brownish grains. The meta-siltstone enclosing the nodules and lenses is generally well cemented (probably silicified), and reddish-purple or gray in color. Many samples have a light-colored zone or *halo* in the siltstone surrounding the epidote meta-calcrite.

Garnet Meta-Calcrite. The garnet meta-calcrite consists of thin lenses or veins of garnet, isolated grains or clots of garnet, and garnet-lined calcite nodules. The garnet veins, which seem to be parallel to bedding planes, are ≤1 mm thick. The nodules are up to a centimeter in diameter. The main minerals present are grandite garnet (usually greenish in color), calcite, chlorite, a clay mineral, plagioclase, and fine-grained reddish-brown hematite. Specular hematite is generally not observed in association with garnet. The rock matrix enclosing the garnet veins and nodules is tan to reddish in color. The reddish samples often have light colored halos, like those described above, at the contact between meta-calcrite and meta-siltstone.

Garnet-Epidote Meta-Calcrite. This type of meta-calcrite occurs as rounded and irregular nodules and patches. The irregularly shaped nodules and patches are fairly large, up to about 4 cm in their longest dimension. Smaller nodules are commonly spherical to ovoid in shape and range from 2 to 10 mm in diameter. Both types of nodules are generally rimmed by fairly coarse-grained garnet (usually reddish-brown in color), and contain calcite, fine- to coarse-grained epidote, fine- to medium-grained garnet, chlorite, and fine-grained reddish hematite. Figure 3 shows an idealized garnet-epidote meta-calcrite nodule. Some nodules have a layer of calcite and/or a fine-grained clay mineral that extends beyond the garnet rim. As with the garnet meta-calcrite, the rock matrix enclosing the nodules may be tan or pale red in color, with halos around nodules occurring in the more reddish meta-siltstone.

CALC-SILICATE MINERALOGY

The most important new minerals formed during contact metamorphism/metasomatism were epidote and grossular-andradite garnet.

Epidote Epidote occurs as fine- to medium-grained, anhedral to subhedral crystals in the matrix of epidote meta-calcrite, where it appears to be a replacement of calcite and clays. Matrix epidotes range from about 10 to 80 μm in length. Coarse prismatic epidote crystals in calcite-filled vugs and in garnet-epidote meta-calcrite nodules are generally euhedral, and seem to have crystallized directly from a fluid. These crystals range in length from about 200 μm to over a millimeter.

Epidote compositions were determined by electron microprobe analysis. Epidote compositions are frequently expressed as the mole percent of the hypothetical end-member molecule *pistacite* (Ps), which is

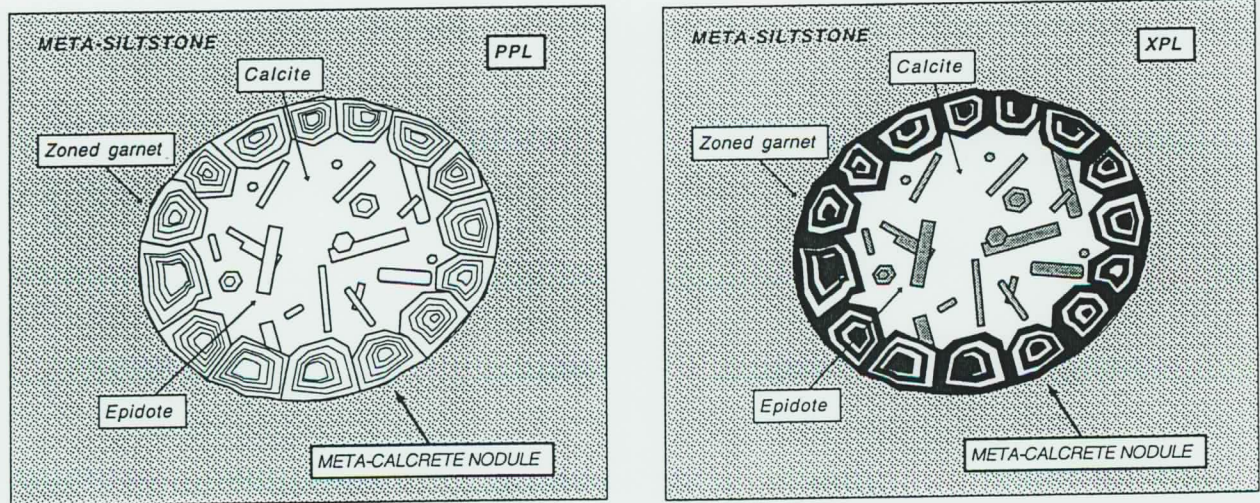


Figure 3. Idealized garnet-epidote meta-calcrete nodule as viewed in plane polarized light (PPL) and cross polarized light (XPL). Nodule is ~ 3 mm wide.

$\text{Ca}_2\text{Fe}_3\text{Si}_3\text{O}_{12}(\text{OH})$. This is calculated as $(\text{Fe}/\text{Fe} + \text{Al}) \times 100$. Epidotes analyzed in this study are moderately iron rich, with $\text{Ps} = 16.9 - 28.5$. (Stoichiometric "epidote," $\text{Ca}_2\text{Al}_2\text{FeSi}_3\text{O}_{12}(\text{OH})$, is $\text{Ps} = 33.3$.) Backscattered electron imaging suggests that some of the epidotes are compositionally zoned, with bright iron-rich cores and darker iron-poor rims. Microprobe analyses of several crystals confirmed this compositional zoning. One such crystal ranged from Ps_{28} to $\text{Ps}_{17.3}$ from core to rim.

Grossular-Andradite Garnet Garnet in the meta-calcrete occurs mainly as euhedral to subhedral crystals, ranging in size from about $5 \mu\text{m}$ to a little over 2 mm in diameter. Most garnets fall into one of two size categories: fine garnets (~ 5 to $20 \mu\text{m}$) and relatively coarse garnets (~ 0.5 to 1.5 mm). In hand sample, garnets fall into two color groups: greenish garnets and dark red-brown garnets. In thin section, some garnets appear isotropic in cross-polarized light, but many grains exhibit anomalous birefringence, which is typical of grandite garnet. The fine garnets are more or less homogeneous in appearance: small pale greenish garnets (clear in plane light) are wholly birefringent, while small brownish garnets (yellow-brown in plane light) are wholly isotropic.

Garnet compositions were determined by electron microprobe analysis. All garnets analyzed are $> 93\%$ grossular-andradite solid solutions. The bulk of the garnet crystals are andradite-rich in composition ($\geq \text{And}_{89}$). Garnet of intermediate or grossular-rich composition occurs only as small ($\leq 30 \mu\text{m}$) grains or as thin zones in larger grains. The small, relatively homogeneous garnets fall into two main compositional groups. The darker-colored, isotropic garnets are close to pure andradite in composition, while the small clear birefringent garnets have more intermediate granditic compositions (And_{47-62}).

The coarse garnets often show signs of concentric (growth) zoning. The zoning is apparent under crossed polarizers, as shown by alternating isotropic and birefringent bands, and may also be evident in plane light, indicated by visible growth surfaces or alternating bands of clear or pale yellow color and darker yellow-brown color. Some coarse greenish garnets are conspicuously zoned, with distinct growth surfaces separating relatively wide ($40-50 \mu\text{m}$) isotropic bands from narrow ($5-10 \mu\text{m}$) birefringent bands. Figure 4(a) shows the results of a microprobe spot traverse of a zoned garnet crystal on a ternary diagram with andradite, grossular and all other garnet end members as the apices. The figure shows that the isotropic zones correspond to andradite-rich compositions, while the birefringent zones have more intermediate grossular-rich compositions. This oscillatory compositional zoning will be discussed in more detail in a later section.

Some unusual garnets consist of a euhedral isotropic core, a thin euhedral birefringent zone or “rim,” and a thick subhedral isotropic “mantle” surrounding the inner euhedral core and rim. Figure 4(b) shows compositions of the isotropic and birefringent zones of these mantled garnets on a ternary plot. Again, the isotropic zones correlate with andradite-rich compositions and the birefringent zones with intermediate granditic compositions.

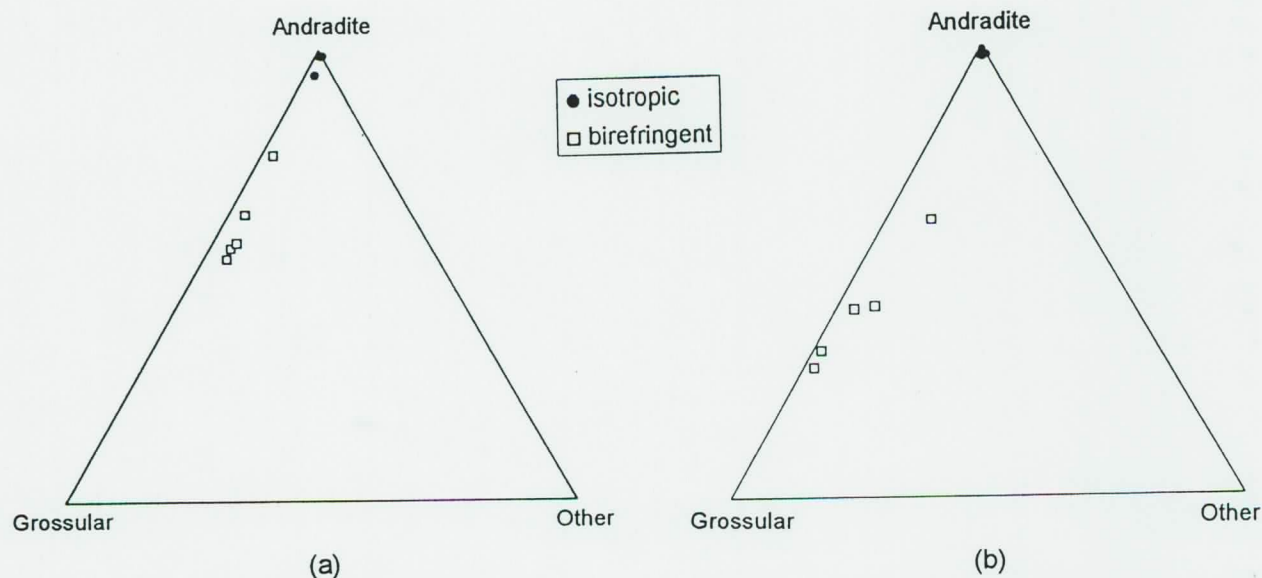


Figure 4. Compositions of isotropic and birefringent zones of (a) Zoned Garnet 1 and (b) Mantled Garnets.

PETROLOGY

Metamorphic Facies, Temperature and Pressure The mineral assemblages of the meta-calcretes suggest that these rocks experienced metamorphic conditions of the albite-epidote hornfels facies, forming at temperatures between about 250° and 450°C and at pressures ≤ 1 kb. Considerations of heat conduction from the Mount Carmel Sill and Dee's Dike, and reported temperatures for active geothermal areas with similar mineral assemblages similarly suggest that the meta-calcretes formed at temperatures between 200° and 400°C. Stratigraphic reconstruction suggests that the meta-calcretes formed at a lithostatic pressure of about 477 bars, since the Mount Carmel Sill is believed to have intruded the New Haven Arkose at a depth of ~ 1800 meters (Toro, 1985; Walton and O'Sullivan, 1950).

Metasomatic Fluids The mineral assemblages, compositions and textures of the meta-calcretes provide information about the metasomatic fluids involved in their formation. This information can be used to make estimates of fluid properties such as fluid pressure (P_{fluid}), f_{O_2} , X_{CO_2} , and pH, and to evaluate the overall role of metasomatic fluids.

Fluid pressure may have varied considerably during contact metamorphism and metasomatism. At times when pore fluids were trapped or isolated, fluid pressure probably equaled lithostatic pressure (P_{lith}), and the fluid would have been in a supercritical state (the critical point of water is 374.1°C and 221.2 bars). As elevated temperatures caused dehydration and decarbonation reactions to occur, volatiles were added to the fluid. Eventually, P_{fluid} may have risen above P_{lith} and exceeded the tensile strength of the rock. The overpressured fluid would have caused hydrofracturing, allowing volatiles to escape and causing a sudden decrease in P_{fluid} . At the

temperatures estimated above, this drop in pressure may have brought the fluid below its critical point, allowing boiling to occur.

The loss of CO₂ during boiling and the lowering of temperature due to evolution of steam would have caused rapid precipitation of calc-silicate minerals. As these minerals filled fractures, CO₂ could have built up in the fluid to the point where calcite precipitated, further sealing the fractures. This would have allowed volatiles to again raise the fluid pressure so that the whole process could have repeated in a cyclical fashion. Such episodic boiling events may have played a major role in producing some of the mineral textures observed, such as fractured grains filled with calcite and garnet crystals that display oscillatory zoning.

The abundance of hematite and the near-complete absence of magnetite in the meta-calcretes suggests that oxygen fugacity was probably greater than that of the hematite-magnetite buffer at the relevant temperature of formation. If the meta-calcretes formed at temperatures between 250° and 400°C, the lower limits for f_{O_2} of formation would have been between 10⁻³³ and 10⁻²³ bars (see Liou *et al.*, 1983). The relatively shallow geologic setting, in sandy sediments perhaps still undergoing compaction (Walton and O'Sullivan, 1950), implies that the metasomatic fluids were water rich. The presence of calc-silicates, and the abundance of epidote in particular, also suggest water-rich fluids with a low activity of CO₂ (Taylor and Liou, 1978; Liou *et al.*, 1985). It follows also that the metasomatic fluids were slightly alkaline, with a relatively high pH (Liou *et al.*, 1985).

Just as pressure may have varied significantly during the contact metamorphic/metasomatic event, it is likely that the f_{O_2} , X_{CO_2} and pH of the fluid also underwent significant fluctuations. The compositional zoning of calc-silicates, and especially the oscillatory zoning of garnets, support this idea (Rose and Burt, 1979; Bird *et al.*, 1984). These fluctuations might have taken place in the following manner.

Both andradite- and epidote-forming mineral reactions are favored by low X_{CO_2} of the fluid phase. However, as these reactions occur, CO₂ is evolved and added to the fluid phase. H₂O is also being produced by the dehydration of clays. If the fluid is trapped in isolated pores and voids, the addition of CO₂ and H₂O would increase P_{Fluid} , eventually exceeding the tensile strength of the rock and causing hydrofracturing. The sudden release of pressure would bring the fluid below its critical point, allowing boiling to occur. During boiling of hydrothermal fluids, H₂ is preferentially partitioned into the vapor phase (Giggenbach, 1980; Yardley *et al.*, 1991), causing oxidation of the remaining liquid (high f_{O_2}). Escape of carbon dioxide from the system would lower X_{CO_2} , consuming calcite and creating andradite and epidote. The enthalpy change associated with boiling of the fluid would lower the fluid temperature, which would also favor rapid mineral precipitation (Philpotts, 1990).

As fractures become filled with rapidly precipitating garnet and epidote, CO₂ would again build up in the system. Eventually, the reactions are reversed and calcite precipitates. Since X_{CO_2} is higher and the pH is lower, the calc-silicates become unstable and calcite + clay + hematite becomes the stable assemblage (Liou *et al.*, 1985). Thus, while andradite and epidote are forming, pH is initially high but is decreasing, and X_{CO_2} is initially low but is increasing. During precipitation of calcite, pH is initially relatively low but is increasing, and X_{CO_2} is initially high but is decreasing; the fluid is reduced to lower values of f_{O_2} as the evolution of steam ceases.

The mineralogical textures observed in the meta-calcretes attest to the importance of metasomatic fluids in forming these rocks. For instance, in the garnet-lined meta-calcrete nodules, the garnets display euhedral crystal faces pointing in toward the center of the nodule while the outer sides of the garnets, in contact with meta-siltstone, are anhedral (see Figure 3). This suggests that the garnets precipitated from fluid filling a void in a manner not unlike that involved in the formation of a geode. The euhedral forms of the epidote crystals within the nodules likewise point to precipitation from a fluid.

Yardley *et al.* (1991) suggested that oscillatory compositional zoning of minerals, such as that displayed by some garnets from Dee's quarry, may be a diagnostic feature of rocks that have undergone infiltration metasomatism. A number of observations suggest that fluid infiltration sometimes took place forcefully, which is consistent with hydrothermal boiling events. Fractured crystals of garnet, with calcite filling the fractures, are common, as are veins filled with calcite, garnet and epidote. Sometimes calcite veins break through the garnet rims of nodules and the vein calcite is obviously continuous with the calcite within the nodule. These features may have been produced when confined, overpressured fluid caused hydrofracturing, and the tensional movements of the surrounding rock caused mineral grains to rupture. The fluid later precipitated calcite, healing the fractures.

Oscillatory Compositional Zoning of Garnet As mentioned above, some garnets from Dee's quarry are composed of alternating birefringent and isotropic growth zones. Backscattered electron imaging of these garnets shows that the zones are compositionally distinct, with sharp boundaries. Microprobe analysis revealed that the wide optically isotropic zones are iron rich while the thin birefringent zones are relatively iron poor. This

oscillatory compositional zoning (or simply oscillatory zoning) is fairly common in grandite garnets from hydrothermal or contact metamorphic/metasomatic environments. Such garnets preserve important information about their crystallization history and about the evolution of the metasomatic fluids from which they crystallized.

For this study, the chemical profile of a relatively large zoned garnet and a smaller mantled garnet were investigated by means of microprobe spot traverses. Zoned Garnet 1 is a euhedral greenish garnet from a specimen of garnet-epidote meta-calcrete. The crystal is $\sim 500 \mu\text{m}$ in diameter and consists of 10 distinct zones. The average composition of the isotropic iron-rich zones is $\text{And}_{97.9}\text{Gr}_{0.9}\text{Other}_{1.2}$ and the average composition of the birefringent relatively aluminum-rich zones is $\text{And}_{62.1}\text{Gr}_{34.4}\text{Other}_{3.5}$. Figure 5 graphically depicts the major and minor element compositions of each zone of the garnet from core to rim. Among the major elements, iron and aluminum oscillate regularly and show an inverse relationship to one another, confirming that most of the iron is present as Fe^{3+} in the octahedral sites of the garnet lattice. Among elements present in minor amounts, manganese and titanium oscillate in sympathy with aluminum. Note that Mn shows an overall increase from core to rim, in both the andradite-rich and grossular-rich zones.

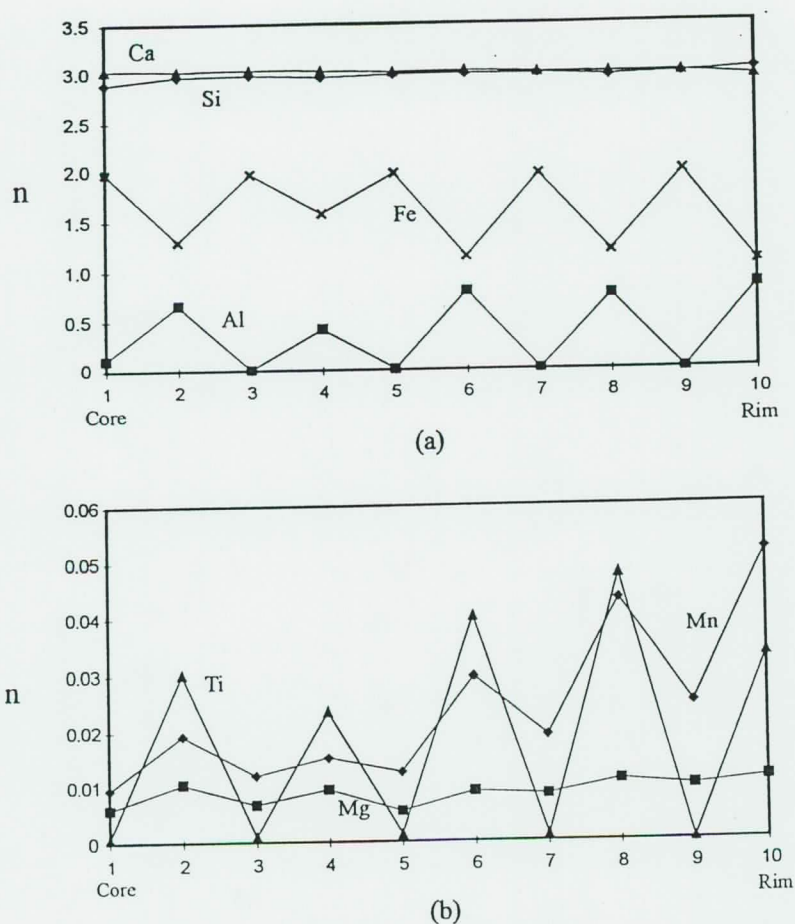


Figure 5. Changing element concentrations in Zoned Garnet 1: (a) major elements; (b) minor elements.

Mantled garnet 1 is a greenish garnet from a sample of garnet-epidote meta-calcrete. The grain is $\sim 200 \mu\text{m}$ in diameter, with conspicuous zoning evident in both plane- and cross-polarized light. Figure 6 provides graphs showing the changing major and minor element compositions of the zones. The zones vary from about

98 % andradite in composition to relatively grossular-rich compositions, up to $\text{Gr}_{66}\text{And}_{32.5}\text{Other}_{1.5}$. Iron and aluminum again exhibit an inverse relationship. Manganese increases steadily rimward, while titanium oscillates along with aluminum.

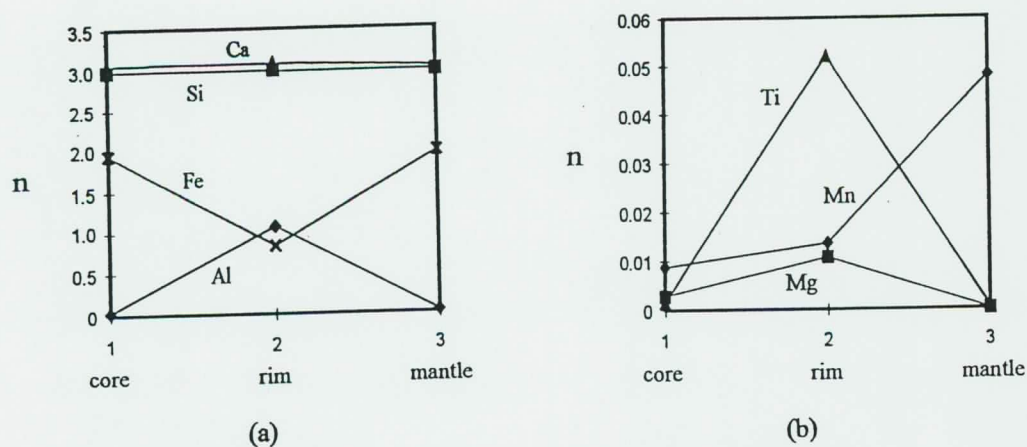


Figure 6. Changing element concentrations in Mantled Garnet 1: (a) major elements; (b) minor elements.

The oscillatory zoning of garnets from Dee's quarry may be explained by cyclical changes in the external conditions of crystallization, such as changes in the composition, temperature or oxidation state of the fluid phase. There is abundant petrographic evidence that episodes of boiling and rapid, forceful fluid flow took place. The fractured garnet and epidote grains attest to these relatively violent events. The presence of different mineral assemblages, such as garnet + epidote + calcite versus calcite + hematite + clay, suggests changes in pressure, X_{CO_2} and pH of the fluid phase such as might be caused by hydrofracturing, pressure release, degassing of CO_2 and boiling of hydrothermal fluids. The distinct growth surfaces present between adjacent zones in some garnets suggest that garnet growth virtually ceased at times, as might be expected at the conclusion of a hydrothermal boiling event. The sequence of thick zones of andraditic garnet rich in Fe^{3+} followed by thin zones of more aluminous garnet suggests periods of rapid crystal growth from oxidized, vigorously circulating fluids, followed by periods of relatively slow growth from more reduced fluids. Again, this is consistent with the occurrence of episodic boiling events (Cf. Yardley et al., 1991 and Jamtveit et al., 1993).

The behavior of manganese, which shows a more or less steady increase in concentration from core to rim in all garnets analyzed, supports the idea of changing external conditions, with fresh batches of fluid of different composition being brought into contact with the growing garnet crystals. Such a rimward increase in Mn concentration is unlikely to occur in garnets precipitated from a single batch of fluid, since garnets preferentially incorporate Mn and typically have decreasing Mn content from core to rim (Hollister, 1966; Yardley, 1989).

The textural differences between the large zoned garnets and the mantled garnets suggest that several generations of garnets may have formed during different episodes of hydrothermal boiling and fluid flushing. The unusual textures of the mantled garnets may be explained by the formation of overgrowths on preexistent cores, in separate boiling events. It is possible that some of the fine-scale oscillatory zoning observed may have resulted from minor, less violent boiling events, involving fluid flashing (in the manner of a geyser eruption) but not hydrofracturing. Such lesser events may have occurred at more closely spaced time intervals while fractures were open and hydrothermal circulation was taking place.

CONCLUSION

Study of the petrography, mineralogy, petrology and geochemistry of contact-metamorphosed calcretes at Mount Carmel, Connecticut, reveals that these rocks represent a fossil geothermal system. The conditions that led to the formation of this system and the skarn-like metasomatic rocks it produced include the intrusion of hot, mafic magma into water-saturated siliciclastic sediments rich in iron oxides and nodules and lenses of soil carbonate. During the existence of the geothermal system, several episodes of hydrothermal boiling were brought on by build-up of volatiles in the fluid phase, which led to hydrofracturing of bedrock followed by a sudden decrease in fluid pressure. These episodic boiling events were instrumental in producing the striking petrographic and geochemical features of the meta-calcretes, namely, the formation of abundant iron-rich calc-silicate minerals, the fracturing of garnet and epidote crystals and meta-calcrete nodules, and the oscillatory compositional zoning of grandite garnet.

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