Chemical disequilibrium during garnet growth: Monte Carlo simulations of natural crystal morphologies

Dru E. Wilbur*
Jay J. Ague† ⎤ Department of Geology and Geophysics, Yale University, P.O. Box 208109, New Haven, Connecticut
⎦ 06520-8109, USA

ABSTRACT

Understanding mineral reaction pathways is critical for determining the pressure-temperature-composition (*P-T-X*) histories of mountain belts. We use Monte Carlo simulations of crystal growth morphologies to assess chemical disequilibrium during deep crustal (25–35 km) garnet crystallization (Connecticut, USA). Initial garnet growth produced crystals with poorly developed crystal faces and/or branched, dendritic forms. This growth phase occurred after equilibrium conditions for devolatilization reactions were considerably overstepped during metamorphic heating as a result of energy barriers to garnet nucleation. Mineral compositions and assemblages may have been unable to track evolving *P-T-X* histories during both the period of overstepping when no reaction was occurring, and when reactions began far from chemical equilibrium. Growth during the overstepped stage must therefore preserve valuable information about the kinetic history of metamorphism, and could have released considerable volatiles, leading to rock weakening and potentially hydrofracturing. The position of regional garnet isograds in the study area may have been strongly dependent on the kinetics of garnet nucleation and growth, rather than the equilibrium *P-T-X* conditions for garnet-forming reactions.

Keywords: garnet, crystal growth, metamorphism, chemical disequilibrium, Monte Carlo simulations.

INTRODUCTION

The chemical compositions of minerals crystallizing in metamorphic rocks are functions of temperature (T), pressure (P), the bulk composition of the rock and interstitial fluids (X), and time. The P-T-X histories of rocks are used to understand a broad spectrum of phenomena, including processes of mountain building and the long-term cycling of elements and greenhouse gases over geologic time scales. These histories are typically quantified assuming that reactions achieve local chemical equilibrium at thin-section scale. However, considerable departures from equilibrium may be much more common than previously thought (Ridley and Thompson, 1986; Rubie, 1998; Spear and Daniel, 2001; Baxter and DePaolo, 2002; Carlson, 2002; Lüttge et al., 2004; Müller et al., 2004). Assessment of such departures has proven to be difficult and controversial, largely because metamorphic rocks preserve complex, time-integrated P-T-X histories spanning time scales of 10^6 yr or more. Nonetheless, a better understanding of the energetics of crystallization is essential because chemical disequilibrium may play a fundamental role in governing the rates of deformation and reaction during mountain building (Baxter and DePaolo, 2004).

SAMPLES AND METHODS

We use chemical maps of garnets and Monte Carlo (MC) simulations of crystal growth to interpret garnet morphologies and assess disequilibrium during garnet growth. The Wepawaug Schist (WepS) and Maltby Lakes metavolcanics (MLM) of Connecticut were metamorphosed from greenschist (~425 °C, ~0.7 GPa) to amphibolite facies (~600 °C, ~1.0 GPa) during the Acadian orogeny (Ague, 2002; Fig. DR1[1]). Dehydration reactions consumed muscovite and chlorite and produced garnet during Barrovian metamorphism of the WepS (van Haren et al., 1996). In most rocks, the garnet cores are anhedral (crystal faces are poorly developed or absent) or have four or more branches in thin section, and are relatively free of inclusions of carbonaceous organic matter (OM) and Fe-Ti oxides (rutile and/or ilmenite) (Fig. 1). Agglomerations of OM on the margins of the cores, numerous inclusions marking crystallographic boundaries, and radially distributed, rod-like inclusions of quartz are also found. The cores give way to rims that contain numerous inclusions

and are characterized by much better developed crystal faces (euhedral). The WepS garnet core morphologies are found elsewhere (e.g., Harker, 1932; Burton, 1986; Jamtveit and Andersen, 1992) and are an exceptionally well developed form of textural sector zoning (TSZ) (cf. Rice and Mitchell, 1991). Similar textures are found in other minerals, including trapiche ruby and emerald (Sunagawa et al., 1999) and andalusite (chiastolite; Rice and Mitchell, 1991). TSZ is not twinning, and it differs from true sector zonation; in the latter, different growth sectors have different chemical compositions. Very rapid growth far from chemical equilibrium has been proposed to explain TSZ (Rast, 1965; Jamtveit and Andersen, 1992), but we develop a new direct test of metamorphic crystallization hypotheses using MC modeling. In the OM-free, greenschist facies meta-ultramafic rocks of the MLM, markedly anhedral, petal-shaped andradite-uvarovite-grossular garnets are found that resemble the cores of some WepS garnets (Fig. 1). Nucleation occurred on relic Cr-rich spinels.

Chemical changes across core-rim boundaries of WepS garnets range from marked (Fig. 2) to subtle (Fig. DR2). The irregularly shaped cores cannot be the result of resorption reactions that consumed initially euhedral garnet because the increases in Mn at the margins of the cores that would have resulted from this process are absent (Kohn and Spear, 2000). The distinct morphologies and compositions of cores relative to rims indicate that cores grew earlier and under different conditions than the rims.

We use the MC method of Xiao et al. (1988) to model crystal growth at constant P (Appendix DR1). As far as we know, this is the first direct application of MC to porphyroblast growth morphologies. The treatment includes diffusion of nutrient through the fluid phase to the growing crystal, surface attachment kinetics, and diffusion along crystal faces. We extend the method to allow for variable amounts of equilibrium overstepping during growth. The simulations are done on a two-dimensional trigonal lattice. Garnet is cubic, but the model retains relevance because the face-centered cubic cell has trigonal packing parallel to {111}. The treatment is also relevant for trapiche ruby (trigonal) and emerald (hexagonal). Nutrient aqueous species dis-

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*Present address: Brinkerhoff Environmental Services, Inc., 1913 Atlantic Avenue, Manasquan, New Jersey 08736, USA.
†E-mail: jay.ague@yale.edu.

[1]GSA Data Repository item 2006139, Figure DR1, location map, Figure DR2, chemical zoning profiles across garnets, Figure DR3, crystal morphology diagram, and Appendix DR1, descriptions of electron microprobe and Monte Carlo procedures, is available online at www.geosociety.org/pubs/ft2006.htm, or on request from editing@geosociety.org or Documents Secretary, GSA, P.O. Box 9140, CO 80301-9140, USA.

Figure 1. Plane polarized light photomicrographs of garnet morphologies. A–E: Garnets from staurolite-kyanite zone of Wepawaug Schist. F: Garnets from meta-ultramafic rocks of Maltby Lakes Metavolcanics. Sample locations are shown in Figure DR1 (see footnote 1).

solved in the fluid are generated in a circular source region at some distance from the growing crystal. Diffusion through the fluid takes place according to a random-walk process through the grid. The treatment recognizes supersaturation, $\Delta\mu/kT$, and normalized bond strength, Φ_1/kT, as important master variables controlling growth morphology (k = Boltzmann constant, T = absolute temperature); $\Delta\mu/kT$ is the thermodynamic driving force for crystallization acting to bond molecular growth units to the crystal. The chemical potential difference $\Delta\mu$ reflects the average Gibbs free energy difference between the compositions of the supersaturated fluid and the fluid in equilibrium with the solid directly at the solution-crystal interface. Crystalliza-

tion at low $\Delta\mu/kT$ near chemical equilibrium favors growth of compact, facetted, euhedral crystals, whereas high $\Delta\mu/kT$ corresponds to strong disequilibrium and results in anhedral or branched forms (cf. Xiao et al., 1988; Sunagawa et al., 1999). The interaction energy, E_i, for a molecular growth unit and its nearest and next-nearest neighbors in the solid is: $E_i = \Phi_1 n_i + \Phi_2 m_i$, where Φ_1 and Φ_2 are the interaction energies per molecule between a given site in the lattice and its n_i nearest neighbors and m_i next-nearest neighbors, respectively. The normalized bond strength Φ_1/kT increases as bond strength increases or T decreases, and ranges from ~4 to ~12 in silicate minerals (Lasaga and Lüttge, 2003); we use an intermediate value of 7 but note that the garnet

morphologies can be reproduced for any value of Φ_1/kT in the likely range for silicates.

An important simplification is that the energetics for only one growth species are considered; individual species (e.g., Fe^{2+}, $H_4SiO_4^o$) in solution are not distinguished. Strictly speaking, this approach is valid for simple minerals such as quartz and trapiche corundum. Full treatment for multisite minerals would include $\Delta\mu$ terms for each site and/or growth species as well as Φ terms for the interaction energies of all the various bonds. These values are not known for garnet, so including them would be premature. We note, however, that at the high P and T of deep crustal metamorphism, Al and Si form complexes (Manning, 2004), and thus the treatment may capture at least some of the basic energetics of molecular-scale Al-Si framework building in garnet. The $\Delta\mu/kT$ and Φ_1/kT can be thought of as average values necessary to assemble growth units composed of several distinct chemical entities. Other model simplifications include neglect of fluid advection and interactions between species in the fluid. Many of these simplifications are similar to those in other recent MC models and should be tested by experiments and ab initio calculations. Nonetheless, MC simulations are useful because they are based on atomic level physics, including the energetics of nearest and next-nearest neighbor interactions, bond strengths, lattice anisotropy, and diffusional transport to the crystal and along crystal surfaces. There are scale-dependent aspects to crystal growth (Xiao et al., 1988), but the general conclusions regarding overstepping and morphology will be appropriate for macroscopic crystals. For example, the transition from branched to compact euhedral crystal forms will shift to slightly smaller $\Delta\mu/kT$ and larger Φ_1/kT as model crystal size increases (Xiao et al., 1990), but growth in the third dimension, which we neglect, tends to have the opposite effect and stabilizes compact euhedral forms at larger Φ_1/kT (Xiao et al., 1991). Thus we infer that these two effects will tend to cancel (Appendix DR1; see footnote 1).

RESULTS AND DISCUSSION

The MC simulations successfully produce model textures that are strikingly similar to the WepS garnet textures (Fig. 3). Growth of the garnet cores must begin in a highly supersaturated (equilibrium overstepped) system. Gradients in fluid composition adjacent to crystals growing in such systems can be considerable, and tend to be largest in areas where multiple crystal faces intersect and kink densities are elevated (cf. Xiao et al., 1988). Because diffusional transport through the fluid
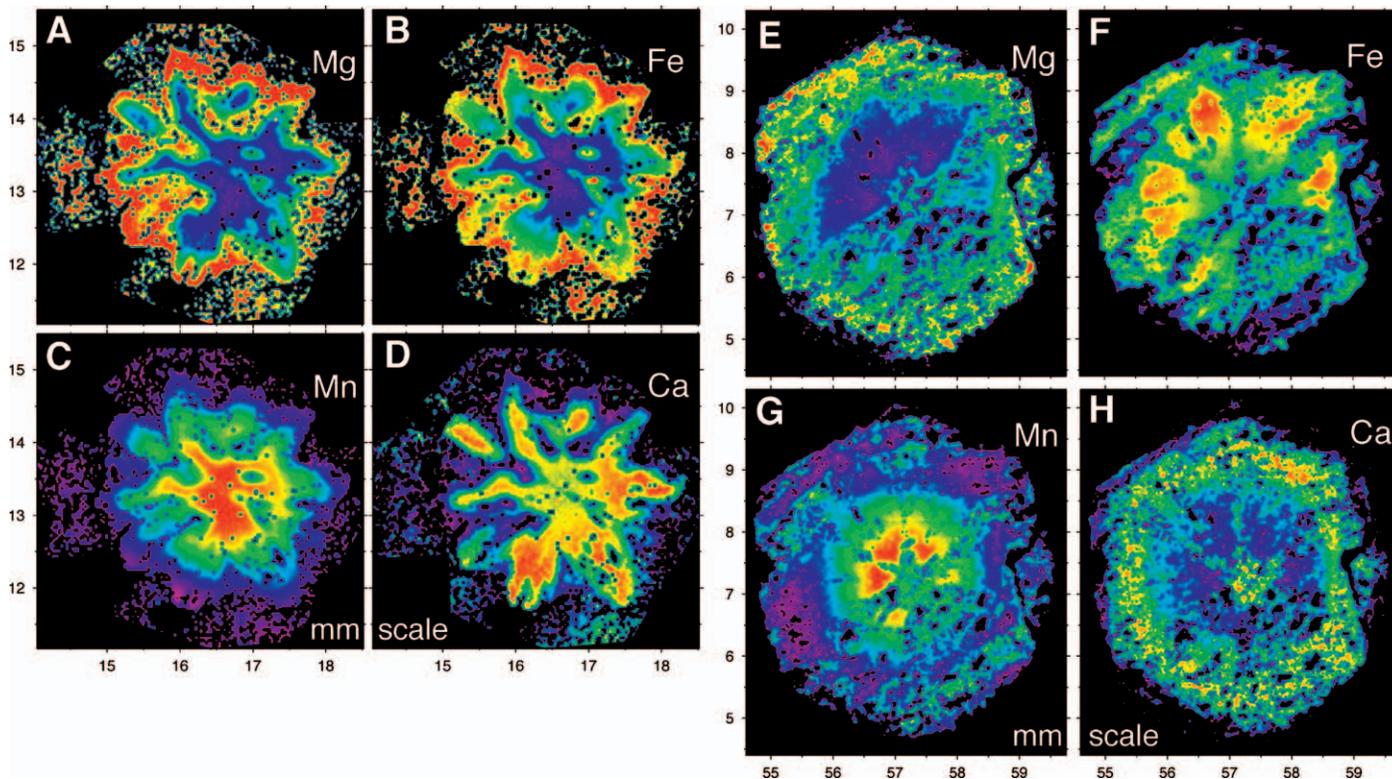
Figure 2. Chemical maps of garnets in Figure 1A (panels A–D) and 1B (panels E–H). Color scales range from purple (lowest concentrations) to red (highest concentrations) (Appendix DR1; see footnote 1). Mn branches in C are shorter than those for other elements (A, B, D), presumably because Mn was depleted from matrix by growing garnets.
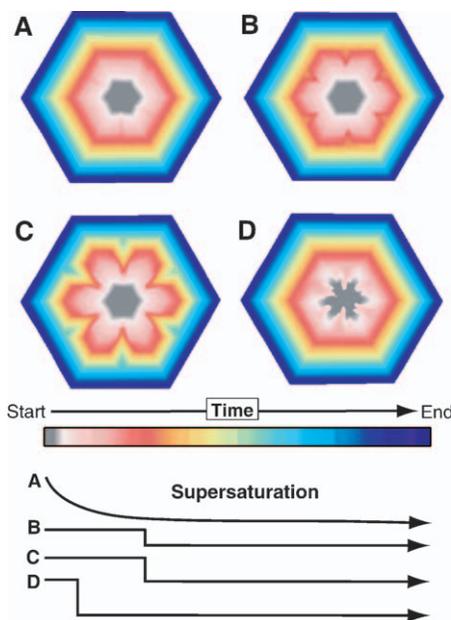
Figure 3. Monte Carlo (MC) crystal growth simulation results (Appendix DR1; see footnote 1). $\Phi_1/kT = 7$; $\Phi_2/\Phi_1 = 0.1$ (see text). A: $Log_{10}(\Delta\mu/kT)$ decreases linearly from 0.7 to -2 during growth. B: $\Delta\mu/kT = 3$ for first 25% of growth history, decreasing to 10^{-2} thereafter. C: $\Delta\mu/kT = 4$ for first 25% of growth history, decreasing to 10^{-2} thereafter. D: $\Delta\mu/kT = 7.5$ for first 7.5% of growth history, decreasing to 10^{-2} thereafter.

is fastest around these areas, they may become morphologically unstable and rapidly grow to project into the surroundings, producing dendritic growth shapes (cf. Shewmon, 1969). As the cores continue to grow, supersaturation is relieved and growth rates slow. Surface kinetics (incorporating surface diffusion and bonding probabilities) then become rate limiting and act to eliminate dendritic perturbations, facilitating the transition to lower-energy, euhedral crystal rims (cf. Xiao et al., 1988; Sunagawa et al., 1999). The minimum value for Φ_1/kT of 4 indicates that increases in Φ_1/kT cannot account for the core-rim textures; consequently, variations in $\Delta\mu/kT$ dominate morphology (Fig. DR3; see footnote 1). Furthermore, Φ_1/kT will tend to decrease as prograde T increases, inconsistent with the observed transitions from branched cores to compact euhedral rims. We envision that significantly overstepped growth also produced the MLM garnets.

Given the MC model simplifications, the estimated minimum overstep of ~2 kJ mol^{-1} needed to produce branched growth for Φ_1/kT of 4 or more (Fig. DR3; see footnote 1) is at best semiquantitative, although it is probably of the correct order of magnitude (similar results have been obtained for square lattices; Temkin, 1966). Oversteps of 1–10 kJ mol^{-1} are consistent with those observed in some

field and experimental studies of geologic dehydration reactions, and translate into equilibrium T oversteps of 40–100 °C (Ridley and Thompson, 1986; Baxter and DePaolo, 2002). Nonetheless, conventional interpretations of metamorphic rocks rarely consider overstepping to be significant.

Hypotheses for crystallization must account for the initial nucleation and growth of the WepS garnets under highly overstepped conditions, and growth at consistently lower supersaturations thereafter. Certain hypotheses, including some type of regional pressure loss or infiltration of chemically exotic fluids, require that the overstepping fortuitously coincided with the initial stages of growth at different times and places across the MLM and WepS, and then never recurred. A simpler hypothesis is that the garnet-producing reaction(s) did not begin at equilibrium P-T-X conditions because garnet nucleation was sluggish during prograde heating. Prior to nucleation, the mineral compositions and assemblages would lag behind changing P, T, and X while the rocks were heated and overstepping increased. Nucleation could have been hindered if the rocks were dry and diffusion rates were slow and approached solid-state rates (cf. Baxter and DePaolo, 2002, 2004). For the MLM, the refractory nature of reactant spinel and product garnet probably hindered

nucleation. Deformation could have increased reactive surface areas and reactivity to promote nucleation, but the triggering mechanism(s) for nucleation remain to be determined. The onset of reaction at elevated $\Delta\mu/kT$ would produce anhedral or branched garnet forms and the associated devolatilization would generate an intergranular pore fluid. Once initiated, overstepped reactions can proceed rapidly; rapid devolatilization would lead to elevated pore fluid pressures, rock weakening, and potentially hydrofracture and even seismogenic failure in tectonically active regions (e.g., Ague et al., 1998). We note that quartz veins in the WepS make up as much as ~30 vol% of the rock mass (Ague, 1994) and probably record brittle failure events.

TSZ development does not appear to correlate with the presence or abundance of OM (Fig. 1). Thus, OM cannot be the sole cause of the TSZ. OM was largely excluded from the WepS garnet cores; the agglomerations of OM on core-rim boundaries may represent the excluded material, but the process of exclusion remains unclear. Fine-grained OM along grain boundaries may reduce grain-to-grain contact areas and/or increase surface free energies between reactant silicate minerals and pore fluids, thus inhibiting nucleation and growth. Increases in grain wetting angles by CO_2 and/or CH_4 (cf. Watson and Brenan, 1987; Holness, 1992) or clogging of pores by OM could reduce rates of transport through intergranular fluids.

We conclude that common garnet growth morphologies preserve important information about chemical disequilibrium in mountain belts. Refinement of crystal growth models will facilitate a better understanding of the kinetics of deep crustal reactions. Mineral compositions and assemblages may have been unable to track *P-T-X* conditions during extended periods of the prograde heating cycle (cf. Baxter and DePaolo, 2002) during overstepping (no reaction) and during the period of growth that produced the anhedral or branched morphologies of the MLM garnets and WepS garnet cores. The rims of the WepS garnets crystallized at lower degrees of supersaturation, so their compositions may be more relevant for equilibrium-based modeling, although considerable disequilibrium is possible even during euhedral growth (Carlson, 2002). Positions of regional isograds may reflect the kinetic timing of garnet nucleation and growth instead of the equilibrium *P-T-X* for garnet formation.

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magnitude. This is perhaps surprising given the differences between the recent open system—possessing a smaller sub-volcanic magma chamber—and the larger-capacity, closed-conduit Plinian system.

To summarize, this work shows that magma recharge in the pre-Plinian system was episodic at the year-to-decade scale and persisted in that state for several decades prior to the Plinian eruption. What is of critical importance to evaluating the volcanic hazard, therefore, is to understand the transition between the magma recharge and buildup stage as investigated here, and the initiation of a Plinian volcanic eruption.

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ERRATUM

Chemical disequilibrium during garnet growth: Monte Carlo simulations of natural crystal morphologies.

Dru E. Wilbur and Jay J. Ague

Geology, v. 34, No. 8, pp. 689–692 (August 2006)

On p. 689, in the first paragraph of "Samples and Methods," Newman, 2001, should have been cited in the sentence describing textural sector zoning. The end of the sentence should read ". . .and are an exceptionally well developed form of textural sector zonation (TSZ) (cf. Rice and Mitchell, 1991; Newman, 2001)." The full reference for Newman (2001) is:

Newman, R.L., 2001, Inter-relationships between metamorphism and deformation during Acadian orogenesis in SW Connecticut [Ph.D. Thesis]: Townsville, Australia, James Cook University, 251 p.