PRESSURE-TEMPERATURE CONSTRAINTS ON THE CRYSTALLIZATION OF THE HARDING PEGMATITE, TAOS COUNTY, NEW MEXICO

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ABSTRACT

A P-T path for the crystallization of the Harding pegmatite magma has been constrained from 1) the experimentally determined liquidus, solidus and phase assemblages for a bulk sample of the pegmatite, 2) isochores for CO₂-H₂O-NaCl fluid inclusions in quartz and beryl from the various lithologic units of the pegmatite, 3) the conditions of metamorphism of the surrounding host-rock, and 4) pegmatite mineral equilibria, including the Li-Al silicates, micas, and feldspars. The magmatic portion of CO₂-H₂O-NaCl fluid inclusions in quartz and beryl from crystallization. If the host-rock temperature at the time of crystallization began at 650°C and 330-350 MPa (11-12 km depth) and continued isobarically to 550°C. Fluid inclusions indicate that the fluid composition (mole %) evolved from 6CO₂:4NaCl to less than 1CO₂:10NaCl during the crystallization. If the host-rock temperature at the time of intrusion was 0 to 200°C below the solidus temperature, then cooling models for a finite sheet indicate that the magmatite crystallization (giant-crystal texture) occurred in 1000 years or less. The subsolidus hydrothermal replacement and re-equilibration continued for several million years. Isobaric cooling continued to 400-300°C, followed by uplift and erosion. Given the estimated age and depth of the pegmatite, the average rate of uplift of the Picuris Range was 0.7-1.0 cm/yr. Li-Be-bearing margarite, prominent in the hanging-wall metasomatic aureole, probably crystallized below the pegmatite solidus. Quartz-spodumene intergrowths exposed in footwall aplite are interpreted as pseudomorphic replacements of early-formed petalite. Eucryptite replacement of spodumene occurred during uplift at 100-200°C.

Keywords: P-T crystallization path, granite pegmatite, regiona metamorphism, Li-Al silicates, petalite pseudomorphs, Li-Be-bearing margarite, fluid inclusions, cooling models, Proterozoic rocks, Harding pegmatite, New Mexico.

INTRODUCTION

The published record of geological studies of the Harding pegmatites and the surrounding Picuris Range in Taos County, New Mexico, spans more than 60 years (see Jahns & Ewing 1976, 1977 for reviews). The main pegmatite displays dramatic internal zoning and is a past producer (1919-1958) of commercial lepidolite, beryl, spodumene and tantalum-niobium minerals. This paper presents a compilation of relevant published data, combined with new field observations, which constrain a pressure – temperature (P-T) path for the crystallization and uplift of the Harding pegmatite magma. We have used 1) the conditions of metamorphism of the
surrounding host-rocks; 2) the experimentally determined liquidus, solidus, and phase assemblages of a bulk sample of the pegmatite; 3) pegmatite mineral equilibria; and 4) isochores for fluid inclusions in beryl and quartz from various lithologic units of the pegmatite.

LOCATION AND GENERAL GEOLOGY

The Harding pegmatite is located in the Picuris Range, 10 km east of Dixon and 30 km southwest of Taos, in sec. 29, T23N, R11E, in Taos County. Proterozoic metamorphic rocks of the Picuris Range are divided, on the basis of contrasting lithologies, into the Ortega and Vadito groups. The debate over the relative ages of these two groups has not ended (Bauer 1984 gives a recent review); however, they probably share similar primary ages of 1750 to 1650 Ma, as neighboring ranges for which geochronological studies have been made (Grambling 1986). The pegmatite intrudes the Vadito Group. The main dike is about 370 m long and up to 80 m wide in outcrop.

Constraints on T and P of Crystallization

Metamorphic equilibria

Geochronological studies (Brookins et al. 1979) and field relations (Long 1974, Jahns & Ewing 1976) indicate that the pegmatite is younger than the nearby plutons and deformation and metamorphism of the surrounding host-rock. The mineral assemblages and textures preserved in the pegmatite and its aureole show no evidence of regional deformation or metamorphism; therefore, the peak pressure experienced during metamorphism of the host rocks provides an upper bound on the pressure of emplacement and crystallization of the pegmatite magma. This is the first constraint that we shall discuss.

Rocks of the Vadito Group, which the Harding pegmatite intrudes, consist of interbedded amphibolites, metavolcaniclastic units, quartz-muscovite-biotite schists, and phyllites. Staurolite and cordierite are abundant in the pelitic units. The Al2SiO5 polymorphs are abundant throughout the Picuris Range; however, in the southernmost Vadito Group rocks, i.e., around the Harding pegmatites, andalusite is found exclusively. Using data on aluminosilicate stability and garnet-biotite geothermometry, Grambling
& Williams (1985) estimated that kyanite-andalusite-sillimanite assemblages in the Pillar - Copper Hill area crystallized at 380 ± 5 MPa and 505 ± 3°C. From an analysis of mineral compositions and stability data for the aluminosilicates, staurolite and chloritoid, Holdaway (1978) estimated that the rocks of the Ortega Group similarly formed at 330 to 370 MPa and 532 ± 20°C. From an analysis of mineral compositions and stability data for the aluminosilicates, staurolite and chloritoid, Holdaway (1978) estimated that the rocks of the Ortega Group similarly formed at 330 to 370 MPa and 532 ± 20°C. These estimates of the metamorphic conditions and relevant metamorphic equilibria for the Picuris range are summarized in Figure 1. Based on the occurrence of cordierite, staurolite and (the aluminosilicates restricted to) andalusite in the vicinity of the Harding pegmatite, we suggest that the peak metamorphic conditions for the Vadito Group rocks in the southern Picuris Range were 340 ± 10 MPa and 550 ± 10°C. The peak metamorphic pressure in the host rocks places an upper bound of 350 MPa for the pressure of emplacement of the magma that led to the Harding pegmatite.

Pegmatite equilibria

Pegmatite magma. The bulk composition of the Harding pegmatite is granitic, as determined by analysis of a representative composite sample, reported in Jahns & Ewing (1976). Splits from this same sample, which represents one of the few good bulk samplings of a complex pegmatite, have been used in several experimental studies to determine the liquidus (Burnham & Jahns 1962), solidus (Vaughan 1963) and phase assemblages (Fenn 1986) as a function of pressure and temperature for a water-saturated Harding pegmatite magma. The results of these experiments have been discussed in Burnham (1979), Burnham & Nekvasil (1986), Fenn (1986), Jahns (1982), Jahns & Burnham (1969), and Luth (1979). The experimentally determined liquidus and solidus are nearly isothermal over the pressure range 200 to 500 MPa. For this range of pressure, the beginning of crystallization is 650 - 675°C, and the beginning of melting is approximately 550°C, making the interval of magmatic crystallization about 100°C (Fig. 2). The phase assemblages with decreasing temperature determined at 500 MPa by Fenn (1986) are remarkably similar to the sequence of zones observed in the pegmatite; compare the description of lithologic units in Table 1 with Figure 1 in Fenn (1986).

Li-Al-silicates. London (1984) has successfully demonstrated the use of the Li-Al-silicate phase diagram to constrain paths of crystallization for Li-rich pegmatites. The principal lithium aluminosilicate minerals in the Harding pegmatite are spodumene and eucryptite. Spodumene occurs in three different lithologic units, with a distinctive habit in each. The bulk of the spodumene occurs in the quartz - lath spodumene zone (see Table 1), where it forms long bladed euhedral crystals (Fig. 3a). The spodumene laths are locally subperpendicular to the hanging-wall contact, and at the base of the unit are arranged in a jackstraw fashion. Many crystals are bent and deformed, and in some exposures appear to wrap around blocks of lepidolite. Inward from the hanging-wall contact, the zonal assemblage spodumene + quartz appears abruptly at the base of the quartz zone. Spodumene is next most abundant in the microcline - spodumene zone, where it occurs as ovoid to irregularly shaped crystals intergrown along their margins with microcline (Table 1, Fig. 4). The third and least common occurrence of
The occurrence and composition of micas in the pegmatite have been described by Lumpkin (1986) and Jahns & Ewing (1976). Muscovite is common in the wall zone, and Li-rich varieties occur in the main quarry area (Fig. 4). The initial crystallization of a Li-Al-silicate in the Harding pegmatite magma appears to have been petalite for a brief period, giving way to spodumene throughout the remaining period of magmatic crystallization. For the magma to become saturated with respect to Li-Al-silicate initially in the petalite field of stability and yet allow spodumene to predominate throughout the remainder of magmatic crystallization further constrains the initial isobaric P-T path to be no more than a few MPa above or below 350.

Micas. The occurrence and composition of micas in the pegmatite have been described by Lumpkin (1986) and Jahns & Ewing (1976). Muscovite is common in the wall zone, and Li-rich varieties occur throughout the other lithologic units. The occurrence of primary green muscovite in the border (rind) and main part of the beryl zone is consistent with pres-
Fig. 3. Primary spodumene occurrences in the Harding pegmatite. a) Quarry wall circa 1944 exposing (from top to bottom) the amphibolite country-rock, the wall (beryl) zone, the quartz zone, and the quartz - lath spodumene zone. b) Detail of the giant-crystal texture of the quartz - lath spodumene zone. White, bladed spodumene crystals (up to 2 m long) are subperpendicular to the hanging-wall contact. c) Spodumene laths arranged in jackstraw fashion in an underground exposure at the base of the quartz - lath spodumene zone (1 to 2 m below the region shown in a). d) Outcrop view of a portion of the microcline - spodumene zone; the distinctive texture ("spotted rock") consists of spodumene orbs in a microcline matrix. e) Thin section photomicrograph (plane-polarized light) of the margin of a spodumene orb (center of orb is to the right of this view) from the microcline - spodumene zone ("spotted rock"). f) Same view (partly crossed polars) showing cross-hatch twinning in the microcline matrix. See text for further discussion.

sure on the order of 330-380 MPa, based on the upper stability of muscovite + quartz and 0.8 < X(H2O) < 1.0 (Fig. 2). In addition, compositions of the primary green muscovite have an average 5 mole % celadonite component (Lumpkin, in prep.), which extends the stability of muscovite in the granite system (Anderson & Rowley 1981). Lepidolite is common in the replacement units and the microcline
Fig. 4. Textures of replaced Li-Al-silicates in the Harding pegmatite. a) Hand sample view of pink spheroids, interpreted to be pseudomorphs after petalite, in albite aplitic matrix from the basal aplitic unit of the main dike. b) The pink spheroids are an intergrowth of spodumene and quartz with sharp boundaries against the albite aplitic, as seen in thin section between crossed polars. c) An enlarged view shows that the spodumene in each spheroid is a continuous single crystal with quartz blebs scattered throughout. Groups of quartz blebs have the same optical orientation, suggesting that they are interconnected in the third dimension. d) Thin section view between crossed polars of eucryptite-quartz intergrowths (dark areas) partly replacing spodumene, set in a matrix of microcline. e) A similar photomicrograph showing spodumene crystals completely replaced by eucryptite + quartz. The latter two samples are from the quartz - lath spodumene zone exposed in the central adit penetrating the main dike.
spodumene zone. Biotite and Li-Be-bearing margarite are restricted to the metasomatic aureole. The occurrence of the Li-Be-bearing margarite is unusual and is being described by Chakoumakos & London (in prep.). For reference, the thermal stability of margarite and margarite + quartz are shown on Figure 2. However, the Li-Be-bearing brittle mica in the Harding aureole is a diociahedral-trioctahedral intermediate, which probably has an enhanced thermal stability compared with ideal margarite. Given the uncertainty of the effects of the Li-for-□, Be-for-Al, and F-for-OH substitutions on the upper thermal stability of Li-Be-bearing margarite, the temperature of formation cannot be constrained; however, crystallization during subsolidus exomorphism would be consistent with an isobaric P-T path near 350 MPa.

**Feldspars.** Feldspars from several lithologic units of the pegmatite have been analyzed for major and minor elements by electron microprobe (Lumpkin 1986). A brief account of this study is given here as it relates to constraints on the P and T path of crystallization of the pegmatite. A detailed study of feldspars of the Harding pegmatite will be presented elsewhere (Lumpkin, in prep.). Two-feldspar pairs in apparent equilibrium were used to calculate temperatures according to the geothermometer of Whitney & Stormer (1977) and assuming a pressure of 300-400 MPa. For T calculations, only those feldspars were chosen that appeared, on the basis of textural observations, to be cogenetic. The temperatures determined for the quartz - lath spodumene zone, microcline - spodumene zone, and cleavelandite replacement units are in the range 325 to 485°C. These temperatures could reflect re-equilibration during pervasive albitionization; however, the uncertainty of whether the feldspar pairs are truly cogenetic does not allow a confident interpretation of these temperatures.

Microcline from the microcline - spodumene zone displays pervasive cross-hatch twinning (see Fig. 3), which implies a monoclinic precursor. The minimum temperature for the monoclinic-to-triclinic inversion in Or-rich alkali feldspars is about 500°C (Brown & Parsons 1989, Ribbe 1983). In the microcline - spodumene zone, perthite bulk compositions of approximately Or₇₀₋₈₀ indicate primary crystallization at T > 550-575°C followed by exsolution of albite below this temperature, followed by the monoclinic-to-triclinic inversion of the Or-rich host at approximately 500°C, and final equilibration at about 350°C based on compositions of host microcline and exsolved albite.

**Fluid inclusions.** Fluid inclusions in quartz and beryl from the various lithologic units of the pegmatite were studied by Cook (1979); a preliminary analysis of these data was reported in Brookins et al. (1979). A revised analysis of Cook's data has been made (Lumpkin & Chakoumakos 1987, Chakoumakos & Lumpkin 1987) and is summarized more fully here. Using the compositions of fluid inclusions as determined by Cook (1979), isochors were calculated with the modified Redlich–Kwong equation of state presented by Bowers & Helgeson (1983a).
For each lithologic unit, Cook determined inclusion compositions to be CO₂ only, CO₂-H₂O mixtures, and CO₂ - H₂O - NaCl-equivalent mixtures. This finding suggests that liquid-vapor immiscibility occurred during crystallization, that different compositions of fluid were trapped at different times during the course of crystallization, or that the inclusions have undergone various degrees of change in size or content since their formation. The calculations of Bowers & Helgeson (1983b) demonstrate that liquid-vapor immiscibility in CO₂-H₂O-NaCl solutions can occur at temperatures as high as 400°C at 300 MPa. However, Sterner & Bodnar (1989) suggested that fluid inclusions in quartz under medium-grade metamorphic conditions may change in size as they re-equilibrate, in response to differential pressure during burial and subsequent uplift. London (1985) has examined fluid inclusions in spodumene + quartz assemblages that form from the breakdown of petalite in the Tanco pegmatite, and found that the P and T inferred from the inclusions in quartz are not consistent with the Li-Al-silicate phase diagram. Given that fluid inclusions in quartz may be suspect and that they constitute the bulk of the inclusions examined in the Harding pegmatite, we can only offer a cautious analysis of the fluid-inclusion data from the Harding pegmatite.

In general, the fluid inclusions indicate that the fluid composition (in mole %) evolved from 6CO₂-4NaCl to less than 1CO₂:10NaCl during the course of crystallization. Isochores for CO₂-H₂O inclusions display a progressive decrease in equilibration temperature from the beryl zone to the quartz - lath spodumene zone to the rose muscovite - cleavelandite unit, consistent with their sequence (wall zone to core) of crystallization or subsolidus re-equilibration. If the CO₂-H₂O and CO₂-H₂O-NaCl inclusions formed under the same conditions simultaneously, then for the various lithologic units the locus of intersections of the isochores for these two compositions should map the progressive change in P and T during crystallization of the pegmatite (Fig. 5). Despite the wide scatter and overlap of fluid densities, a progressive decrease in formation temperature following an approximately isobaric path is apparent for the primary lithologic units. This proposal is consistent with their inferred sequence of crystallization; however, the pressure indicated is

![Fig. 6. Pressure - temperature path (bold curve) for the magmatic crystallization (solid line), hydrothermal stage (short-dashed line) and uplift (long-dashed curve) of the Harding pegmatite. Selected equilibria from Figures 1 and 2 also are shown and include the aluminosilicate phase diagram (solid lines), the Li-Al-silicate phase diagram (bold solid lines), and the liquidus (short-dashed line) and solidus (long-dashed line) of the water-saturated Harding pegmatite magma. The region of magmatic crystallization is shaded. See Figures 1 and 2 for references.](http://pubs.geoscienceworld.org/canmin/article-pdf/3446217/287.pdf)
100–200 MPa lower than that consistent with the Li-Al-silicate phase diagram. See London (1985) for a similar appraisal of Cook's original findings.

DISCUSSION

The predominant Li-Al-silicate in the Harding pegmatite is spodumene, but there is textural evidence of early-formed petalite. Its former presence suggests that the pegmatite magma must have become saturated with respect to Li-Al-silicate at a pressure slightly below or at the phase boundary between spodumene + quartz and petalite + quartz (Fig. 2). Furthermore, this pressure range of 330–350 MPa is consistent with the upper bound of 350 MPa provided by the peak regional metamorphic conditions. Because the pegmatite liquidus and solidus are nearly isothermal over the pressure range 330–350 MPa (11–12 km depth), the magmatic portion of crystallization began at ~ 650°C and continued isobarically to ~ 550°C (Fig. 6). The host-rock temperature at the time of intrusion could have been anywhere between 0 and 200°C below the pegmatite solidus temperature (i.e., the temperature of the host rock must have been between the peak metamorphic conditions of 550°C and 350°C, the temperature at 11–12 km for a normal geothermal gradient of 30°C/km). Therefore, isobaric cooling through at least the magmatic stage is a reasonable assumption.

Given the available information, we can examine the cooling history of the Harding pegmatite magma using established mathematical theory (Lovering 1935, 1936, Carslaw & Jaeger 1959, Jaeger 1964). Of interest are the maximum temperature
experienced by the aureole, and the time required for the magma body to thermally equilibrate with the host rock. As noted by Jaeger (1964), a realistic theory requires detailed knowledge about the intrusion (emplacement rate, geometry and dimensions of the magma chamber, depth of cover), the magma (initial temperature, thermal diffusivity, heat of crystallization, temperature range of crystallization), and the host rock (initial temperature, thermal diffusivity, geothermal gradient). Further complications include the thermal effects of volatiles in the host rock, of volatiles or convection in the magma, heterogeneous texture or differentiation during the progress of crystallization, and heat production due to radioactivity. Much of the necessary information has been established in the previous section or can be reasonably estimated. A simplified model has been chosen to illustrate the important general features. Consider the cooling of a parallelepiped body of magma of width $2d$ and length $2l$ at a depth $h_2$ below the surface and with its lower surface at depth $h_1$. If the magma, initially at temperature $T_0$, is injected into country rock at temperature $T_e$, the temperature at any position $(x, y, z)$ at time $t$ is

$$T(x,y,z,t) = T_e + \frac{1}{2}(T_0 - T_e) \left\{ \text{erf}\left(\frac{x+d}{2(xt)^{1/2}}\right) - \text{erf}\left(\frac{x-d}{2(xt)^{1/2}}\right) \right\} \left\{ \text{erf}\left(\frac{y+h}{2(yt)^{1/2}}\right) - \text{erf}\left(\frac{y-h}{2(yt)^{1/2}}\right) \right\} + \text{erf}\left(\frac{z+h_2}{2(zt)^{1/2}}\right) - \text{erf}\left(\frac{z+h_1}{2(zt)^{1/2}}\right)$$

(Carslaw & Jaeger 1959). Underlying assumptions are that the thermal diffusivities ($\alpha$) of the magma, subsolidus assemblage, and the host rock are the same, and that the thermal contribution from the heat of crystallization is negligible.

First, assume instantaneous emplacement of the pegmatite magma at the temperature of the liquidus, 650°C. At the beginning of crystallization, the igneous body is a parallelepiped with dimensions $2 \times 2 \times 0.02$ km and oriented such that this finite sheet is parallel with the surface. The depth to the upper surface of the body is 11 km. The thermal diffusivity is assumed to be everywhere the same at 0.01 cm²/s. Cooling curves for the center of the pegmatite and positions within the aureole are shown in Figure 7 for a host-rock temperature of 350°C. The magma cools quickly while the contact heats rapidly to a maximum of $\frac{1}{2}(T_1 - T_0)$. Any point in the aureole heats to a maximum temperature (diminishing with distance) and then cools gradually. For higher host-rock temperatures, the general character of the cooling curves is the same. For host-rock temperatures below 550°C, the magmatic crystallization (giant-crystal texture) would have occurred in 100 years or less. For a host-rock temperature near the pegmatite solidus (550°C), the magmatic portion of crystallization would have taken much longer (> 1000 years).

The subsolidus hydrothermal replacement and re-equilibration (i.e., “stewing in its juices”; Jahns 1982) continued for several million years. Nearly isobaric cooling continued to 400–300°C, followed by uplift and erosion (Fig. 6). The decompression is shown to follow a 30°C/km geotherm, although the average slope of the fluid-inclusion isochors might indicate an initially greater change in pressure for the uplift trajectory. Given the estimated age and depth of the pegmatite, the average uplift rate of the Picuris Range is inferred to have been 0.7 – 1.0 cm/yr.

Figure 8 summarizes the crystallization sequence for the important minerals of the Harding pegmatite and its aureole. Early in the magmatic stage, major concentrations of beryl and columbite-tantalite crystallized in the wall zone at 650 – 625°C. Quartz-spodumene intergrowths exposed in footwall aplite are interpreted as pseudomorphic replacements of early-formed petalite. Microlite crystallized in association with spodumene late in the magmatic stage at 575 – 550°C. Additional microlite formed during the hydrothermal stage, as extensive masses of cleavelandite, muscovite and lepidolite replaced pre-existing pegmatite. Pseudomorphs of rose muscovite after spodumene laths and casts of beryl crystals in lepidolite masses are irrefutable evidence that previously solid pegmatite has been replaced (Chakoumakos 1978). Li-Be-bearing margarite, prominent in the hanging-wall metasomatic aureole, probably crystallized below the pegmatite solidus. In support of this contention, London (1986) noted that the mineral assemblages of the metasomatic aureole are characteristic of greenschist-facies metamorphic conditions (i.e., temperatures); thus the infiltration of

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Fig. 8. Crystallization sequence of major and minor minerals in the Harding pegmatite and its metasomatic aureole. The magmatic, hydrothermal, and uplift stages of crystallization correspond to the crystallization path shown in Figure 6. Minerals marked with an asterisk occur only in the metasomatic aureole.
pegmatitic fluids into the host rock that gave rise to the aureole must have occurred at subsolidus temperatures. The inferred P-T path in Figure 6 suggests that most of the eucryptite replacement of spodumene occurred during uplift at 100-200°C.

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