Three-dimensional observation of the boundary region between massive feldspar and graphic granite by X-ray computed tomography

Susumu IKEDA**, Yoshito NAKASHIMA*** and Tsukasa NAKANO***

**WPI-Advanced Institute for Materials Research (WPI-AIMR), Tohoku University, Sendai 980-8577, Japan
***Institute of Mineralogy, Petrology and Economic Geology, Graduate School of Science, Tohoku University, Sendai 980-8578, Japan
***Geological Survey of Japan, National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba 305-8567, Japan

The three-dimensional (3-D) structure of crystals around the interface between massive potassium (K)-feldspar and quartz-feldspar intergrowth (graphic granite) of two samples collected from a pegmatite body in Ishikawa town, Fukushima Prefecture, Japan, was determined by X-ray computed tomography (CT). Based on the results and additional information obtained by serial thin section and polarizing observations, the formation processes of graphic granite and pegmatite bodies are discussed. The most important finding of the 3-D observation using X-ray CT is the very flat spatial distribution of the tips of the quartz crystals. The sharp interface between the graphic and single-phase parts suggests that the change from graphic granite to massive K-feldspar crystallization corresponds to the transition of the thermodynamic conditions at the growth front from non-equilibrium to equilibrium. This indication leads to a new model for the pegmatite genesis, that is, graphic granite forms at the marginal zone under non-equilibrium conditions shortly after the intrusion of magma due to rapid cooling by the cold host rock body and, subsequently, the crystallization of massive K-feldspar starts when the conditions at the crystal growth front reenter the ‘feldspar + melt’ field in the phase diagram due to the relaxation of the conditions. Other results related to the conditions of the growth environment with coexisting aqueous fluid, such as the connectivity of the quartz crystals, are also discussed, for example, the influence of the supercritical state on the nucleation of quartz crystals.

Keywords: Graphic granite, Pegmatite, X-ray computed tomography, 3-D analysis, Thin section

INTRODUCTION

Granitic pegmatites have been of geological interest for more than a century (e.g., London and Kontak, 2012). Giant crystals and noble minerals attract much attention as targets of industrial mining, mineralogical and petrological research, and mineral collections. The pegmatite genesis had been one of the mysteries until the first half of the 20th century, although some important mineralogical findings had been reported (e.g., Fersmann, 1929). One of the milestones in the study of pegmatites was reached by Jahns and Burnham (1969). They proposed a genetic model for pegmatites based on the careful consideration of results of field and laboratory studies. Most geologists and mineralogists today are qualitatively accepting their proposed landmark model based on which aqueous fluid that accumulated by the crystallization of anhydrous minerals in granitic plutons and/or other volatile substances (fluxes) enriched in residual melts plays important roles in the formation of the characteristic features of pegmatites including the growth of giant crystals. However, the details of the genesis are still under debate (e.g., London, 2005; Simmons and Webber, 2008; London and Kontak, 2012; London and Morgan, 2012).

One of the final goals of pegmatite studies is to tell the whole story of the pegmatite formation, that is, how giant crystals grow, how similar zonal (concentric or layered) structures form, and how special textures of minerals, such as graphic textures, which are not observed in...
usual granitic rocks, are produced. Although every pegmatite body has a unique structure, massive feldspar and quartz occupy the center and inner periphery and graphic granite is observed in the outer periphery (marginal zone) of the pegmatite bodies in many cases (e.g., London, 2009). Because the complete experimental reproduction of pegmatite is difficult due to size and timescale limitations, we have to consider this complex issue by collecting enough information from various viewpoints, field observations, and experiments. A lot of experimental work has been carried out to understand the formation of pegmatites (e.g., Fenn, 1986; London et al., 1989; Baker and Freda, 2001). Many of the studies focused on the textural reproduction of the graphic textures, that is, quartz-feldspar intergrowth. Rocks with such a graphic texture in granitic pegmatites is called ‘graphic granite’. Graphic granite consists of giant single-crystalline alkali feldspar (typically, microcline–microperthite), which contains quartz rods with cross-sectional patterns such as gothic characteristics (graphic texture). The size of each quartz rod is on the centimeter scale and single blocks of graphic granite sometimes exceed 1 m. London and Morgan (2012) pointed out that graphic granite defines the unique texture of pegmatites and strongly suggested that graphic granite is the key to explaining the genesis of pegmatites. Graphic granite provides much information about the dynamic process of the pegmatite formation.

The central question discussed with respect to the formation mechanism of graphic granite is whether the texture of the quartz-feldspar intergrowth is due to ‘eutectic crystallization’, which is often observed in metallic binary or ternary systems. Because the texture of graphic granite resembles eutectic textures, it is reasonable that eutectic crystallization is involved in the formation of graphic granite. In fact, such a graphic texture of quartz and feldspar has been reproduced around the eutectic point in some experiments, for example, in those of Baker and Freda (2001) who used starting material with a chemical composition almost the same as that of the eutectic point. However, Barker (1970) strongly suggested that the graphic texture is not the result of simple eutectic crystallization but the result of simultaneous crystallization of quartz and feldspar in the presence of an aqueous fluid. The major reason why Barker (1970) denied the eutectic crystallization as the mechanism was that the bulk chemical compositions of graphic granite is not clearly related with any known eutectic point or cotectic curves between quartz and alkali feldspars. It is difficult to achieve eutectic crystallization if the bulk composition is far from the eutectic point. Fenn (1986) concluded from his experimental results that the graphic texture is not caused by eutectic crystallization but produced by the simultaneous crystallization of quartz and feldspar under kinetically driven non-equilibrium conditions. Ambiguity lies in the fact that eutectic–like graphic textures are observed in natural pegmatites, although their bulk chemical compositions differ from that of the eutectic point.

To provide other viewpoints and constraints on this problem, we investigated the boundary region between massive potassium (K)–feldspar and quartz–feldspar intergrowth (the endpoint of the intergrowth) in natural graphic granite samples by X-ray computed tomography (CT) in this study. We infer that some characteristic features remain at the endpoint (forefront) of the intergrowth because the conditions that enable simultaneous crystallization change at this interface. We conclude that three-dimensional (3-D) observations using X-ray CT are among the best tools to study the structure of such intergrowth endpoints. Based on X-ray CT, we can obtain important information such as the shape, positional relation, and interconnection of crystals which are easily missed by using two-dimensional (2-D) observations only (Ikeda et al., 2000).

**EXPERIMENTAL**

**Graphic granite samples**

We surveyed the Ishikawayama mine (exploited and mined by the Mazak Stone Corporation) in Ishikawa town in the Shionohira District, Fukushima Prefecture, Japan (37.1668°N, 140.4324°E) and collected rock samples in cooperation with the Mazak Stone Corporation. Many granitic pegmatites can be found in the area around Ishikawa town (Kondo, 1953; Matsubara, 1956; Mutsu-mori et al., 2000). This area is part of the largest granitic rock regions in Japan, which is called Abukuma granite (Abukuma Plateau). The K–Ar age of the surrounding graphic granite body was estimated to be 90–120 m.y. (Kawano and Ueda, 1967; Shibata and Uchiimi, 1983). Similar age values were obtained using the Sm-Nd and Rb-Sr methods (Shibata and Tanaka, 1987). The Ishikawayama mine is a pegmatite mine with roughly ellipsoidal shape (the longest and shortest diameters of the ellipsoid are ~30 and 10 m, respectively) and a zonal mineral species structure. The outer periphery (marginal zone) of the ellipsoidal body mainly comprises graphic granite (up to ~5 m in thickness). Tourmaline crystals are frequently observed in this graphic granite layer. Toward the center of the body, the graphic granite zone is followed by a massive K–feldspar zone around the middle periphery. The inner periphery and center of the body are occupied by giant crystals of quartz and K–feldspar. The specimens used in this study were collected from a boundary part,
where the graphic granite zone transfers into the massive K-feldspar zone. The graphic granite is mainly observed in the outer part of the ellipsoidal pegmatite body (e.g., Kondo, 1953; Jahns and Burnham, 1969; Mitsumori et al., 2000; London, 2009); the quartz in the outer part of the ellipsoidal pegmatite body (e.g., Kondo, 1953) where the graphic granite zone transfers into the massive K-feldspar seem to start at the periphery and proceeds toward the interior of the pegmatite body. The quartz rods disappear at a certain point and are followed by massive K-feldspar. The two samples shown in Figure 1, which include the interface between massive K-feldspar (microcline microperthite) and graphic granite, were used for 3-D X-ray CT observations. Sample 1 (Fig. 1a) was cut into a rectangular parallelepiped and Sample 2 (Fig. 1b) was cut and ground into cylindrical shape. In both samples, the K-feldspars in the graphic granite and massive feldspars are part of a continuous single crystal, without the change of the crystallographic orientation. Based on the assumption that the pegmatite formation (crystallization) proceeded from the outer part (marginal zone, which is in contact with the surrounding host granite body) to the inner part (inner periphery and center of the pegmatite body), the quartz-feldspar intergrowth shown in Figure 1 presumably formed (crystallized) from the upper part of the samples and ended at the interface with the massive K-feldspar.

**X-ray CT measurement**

The X-ray CT was first invented for medical use (Hounsfield, 1973) and has also become a standard tool for 3-D observations in Earth and planetary sciences (Wellington and Vineger, 1987; Nakano et al., 1992; Denison et al., 1997; Denison and Carlson, 1997; Nakano et al., 1997; Nakashima et al., 1997; Brown et al., 1999; Uesugi et al., 1999; Ikeda et al., 2000; Nakano et al., 2000; Ketcham and Carlson, 2001; Tsuchiyama et al., 2001; Uesugi et al., 2001; Ikeda et al., 2004; Nakashima et al., 2004; Tsuchiyama et al., 2005; Jerram and Higgins, 2007; Ebel and Rivers, 2007; Remeysen and Swennen, 2008; Nakashima et al., 2008; Okumura et al., 2008; Nakamura et al., 2008; Tsuchiyama et al., 2014; Ikeda et al., 2017). The X-ray CT is a fundamental technique that can be used to measure the spatial distribution of LACs, the realistic output value depends on the system and is called ‘CT value’, which is sometimes an absolute LAC and sometimes a dimensionless relative value. Many medical scanners provide the dimensionless CT value called ‘Hounsfield CT number’ or ‘Hounsfield Unit (H.U.)’, which is normalized using the linear attenuation coefficient of water, as defined by the following equation,

\[
\text{CT value (Hounsfield Unit : H.U.)} = \frac{\mu - \mu_w}{\mu_w} \times 1000 \tag{2}
\]

where \(\mu_w\) is the LAC of water. In the case of graphic granite, the CT values (H.U.) of K-feldspar and quartz are \(\approx 2080\) and \(\approx 1980\), respectively, at 120 kV.

**Beam-hardening correction**

Because medical CT scanners use a continuous X-ray beam, the reconstructed images (2-D slices) are influenced by beam-hardening (e.g., Denison et al., 1997). The low-energy component of the X-ray is more absorbent, thus most of the low-energy photons are absorbed in the periphery of a specimen shortly after entering the specimen. Therefore, the CT value increases at the periphery of the specimen, even if the specimen is homogeneous.

In this study, we corrected this beam-hardening artifact based on the method reported in Nakano et al. (2000). As explained in detail in the Results section, the beam-hardening artifact was removed by subtracting
After beam-hardening correction, segmentation (extraction of the parts of interest by thresholding) was carried out to extract the quartz crystals from the CT images. The threshold values were determined based on a comparison between the CT and thin section images obtained by putting the thin sections between two polarizing plates. This digital image processing produces binary images and enables us to analyze the 3-D structures.

In our previous study (Ikeda et al., 2000), we focused on the 3-D connection of quartz crystals in graphic granite. Therefore, as the first analysis step, we investi-
gated the 3-D connection in this study using the same ‘cluster labeling method’, which is based on the percolation theory (Stauffer, 1985; Nakano and Fujii, 1991). The connection of the voxels in quartz was judged based on the same rule as that used in Ikeda et al. (2000). We call the connected voxels ‘cluster’. The degree of quartz rod interconnection was quantitatively assessed using the value called ‘connectivity’. This value is the volume fraction of the largest quartz cluster to all quartz crystals and defined as follows (Nakano and Fujii, 1989):

\[
\text{Connectivity} = \frac{\text{Volume of the largest cluster}}{\text{Total volume of all quartz crystals}} \quad (3).
\]

**Thin sections**

After the X-ray CT measurements, the specimens were cut every 5.0 mm parallel to the slices of the X-ray CT images and thin sections of the planes were produced. The positions of the thin sections in the samples are shown in Figure 2. Because the thin section and crystal sizes are large, optical observations were carried out by placing the thin sections between two large polarizing plates (crossed polars). Photographs were taken by putting the set of thin sections and polarizing plates on a light table (without a polarizing microscope).

**RESULTS**

**Corrected images of X-ray CT**

We obtained X-ray CT images of the two samples. The raw images (CT images obtained by reconstruction calculations using projection images acquired from various rotation angles) show the beam-hardening artifact, which was corrected using the method reported in Nakano et al. (2000). Figure 3 shows the correction process. Figure 3a shows one slice of the raw CT image of Sample 1 (slice no. 40). Figure 3b shows the simulated image corresponding to the slice shown in Figure 3a. To create the simulated image, the 2-D cross-sectional shape of the sample, average chemical composition of the sample (slice), density of the sample (slice), and spectrum of the incident X-ray beam are needed. The average chemical composition and density are required to calculate the average LAC of the slice (McCullough, 1975; Hubbell and Seltzer, 1996). This simulation technique has also been demonstrated in Nakashima and Nakano (2012, 2014). The mass attenuation coefficient (MAC; \(\tau [\text{cm}^2/\text{g}]\)) is another type of absorption coefficients, which is fundamentally determined by the chemical composition and X-ray energy. However, the density (\(\rho\)) of the substance is needed to calculate LACs:

\[
\mu [\text{cm}^{-1}] = \tau [\text{cm}^2/\text{g}] \times \rho [\text{g/cm}^3] \quad (4).
\]

Therefore, we calculated the MACs before the estimation of LACs and the LACs were obtained by multiplying the MACs with the estimated density. Based on the sample shape, sample size, LACs for used energies, and X-ray spectrum shown in Figure 3d (Nakano et al., 2000), the simulated image of the average attenuation of the slice was created. Figure 3c shows the corrected image obtained by subtracting Figure 3b from Figure 3a. Figure 3e shows the line profiles of the simulated (theoretical) and actual (observed) CT values along the dotted line shown in Figures 3a-c. Because the graphic granite samples are mainly composed of K-feldspar, the simulated profile (simple curve) is almost consistent with the observed profile, while the two profiles differ with respect to the quartz crystals.

Based on the data correction explained above, X-ray CT images without the beam-hardening artifact could be obtained, as shown in Figures 4 (Sample 1) and 5 (Sample 2).

**Connection analysis of quartz**

In our previous study (Ikeda et al., 2000), we showed the great potential of X-ray CT as a tool to analyze the internal connection of crystals in rocks such as quartz rods in graphic granite. In this study, we first applied connection analysis (cluster labeling) to the X-ray CT data of two graphic granite samples. The connectivity values defined by Eq. (3) are \(\sim 0.64\) for Sample 1 and \(\sim 0.95\) for Sample 2 when 54 CT slices are used. These connectivity values increase with increasing analyzed volume until the analyzed volume size reaches the average size of the connected clusters. Thus, the actual connectivity must be larger than 0.64, even in Sample 1. These high connectivity values suggest that most quartz crystals are connected until the very end of the quartz-feldspar intergrowth, similar to the inner part of the graphic granite observed in Ikeda et al. (2000).

**Polarizing observations on thin sections**

The results of the polarizing observations on the thin sections are shown in Figures 6 and 7 together with the corresponding X-ray CT slices. The K-feldspar shows full extinction in all sections, indicating its single-crystalline nature, although it has a perthite structure (microcline-
microperthite) on the microscopic level. Sample 1 shows the almost complete extinction of quartz crystals. This means that most of the quartz rods have a uniform crystallographic orientation, that is, they are single-crystals. On the other hand, Sample 2 shows a smaller degree of extinction; only some groups of quartz crystals show extinction in Sample 2. Interestingly, there are some cases in which quartz crystals do not show extinction in Sample 2, although they are completely connected.

**Characteristic morphological feature of the edge of the quartz-feldspar intergrowth**

Figure 8 shows 3-D images of the endpoints of quartz rods viewed from different azimuthal angles. These 3-D views clearly show the sharp interface between the graphic and single-phase parts of massive K-feldspar. This indicates that the crystal growth of all quartz rods ends almost on the same plane (level).

---

**Figure 3.** An example of the correction of the beam-hardening artifact. (a) Observed (ordinarily reconstructed) image. (b) Simulation image based on the X-ray energy distribution shown in (d), average chemical composition, and density of the sample (K-feldspar and quartz). (c) Corrected X-ray CT image created by the subtraction of the simulated image (intensity) (b) from the raw X-ray CT image (a). (d) Distribution profile of X-ray energy of the X-ray CT scanner (Nakano et al., 2000). (e) Line profiles showing the theoretical and observed curves and difference between them along the dotted lines of (a) to (c).
Figure 4. (a) X-ray CT images of Sample 1 after the correction of the beam-hardening artifact. The size of each image is 135 × 135 pixels (3.6 × 3.6 cm²). The numbers in the individual images indicate the slice number corresponding to the numbers shown in Figure 2. The gray scale indicates the 16-bit CT value and the number in parentheses is the corresponding Hounsfield CT number (Hounsfield Unit; H.U.). Bright parts have large CT values. (b) 3-D view made by stacking the 54 slices.
Figure 5. (a) X-ray CT images of Sample 2 after the correction of the beam-hardening artifact. The size of each image is 160 × 160 pixels (4.3 × 4.3 cm²). The numbers in the individual images indicate the slice number corresponding to the numbers shown in Figure 2. The gray scale indicates the 16bit CT value and the number in parentheses is the corresponding Hounsfield CT number (Hounsfield Unit; H.U.). Bright parts have large CT values. (b) 3-D view made by stacking the 54 slices.
Figure 6. Photos of the thin sections of Sample 1 taken under crossed polars (at the extinction position of quartz crystals and the diagonal position). The corresponding X-ray CT slices are also shown.
Figure 7. Photos of the thin sections of Sample 2 taken under crossed polars (first the polarizing plates were set to a certain angle and then the plates were rotated by 45°). The corresponding X-ray CT slices are also shown. The circled quartz crystals in the photos of thin section no. 4 do not show simultaneous extinction, although they are completely connected.
DISCUSSION

Quartz connectivity and single-crystalline degree

As shown in our previous paper (Ikeda et al., 2000), the 3-D connection of quartz crystals is one of the most interesting characteristics of graphic granite. The possibility of a 3-D connection had been suggested first by Simpson (1962) based on the successive grinding of a graphic granite specimen. It was confirmed by Ikeda et al. (2000)

Figure 8. 3-D images of quartz rods in Samples 1 and 2 viewed from the side. The 3-D images were constructed using ImageJ software (U.S. National Institutes of Health; http://imagej.nih.gov/ij/).
by X-ray CT observations and 3-D image analysis. Such a 3-D connection explains the simultaneous extinction of the quartz crystals in polarizing observations. The quartz crystals in graphic granite seem to be separated in 2-D cross-sections, but they show the same extinction, like a single crystal. The most rational explanation of the simultaneous extinction is that quartz formed a 3-D network (connected) structure by branching and kept a single-crystal state during the formation of graphic granite.

In this study, a difference in the simultaneous extinction (single-crystalline degree) was observed between Samples 1 and 2, although the cluster labeling analysis suggests a large connectivity for both samples. The quartz crystals in Sample 1 (Fig. 6) show almost complete simultaneous extinction in the polarizing observations, which means that the quartz crystals have the same crystallographic orientation in the entire specimen. On the other hand, some groups of quartz crystals in Sample 2 (Fig. 7) show simultaneous extinction at different azimuthal angles and some quartz crystals do not show simultaneous extinction, although they are completely connected (Sample 2 in Fig. 7). This means that the 3-D network of quartz in Sample 2 comprises several groups of branches (the crystallographic orientation is the same within each group but different between the groups), although they are connected in three dimensions.

The following two hypotheses possibly explain the connection (contact) of the quartz crystals with different crystallographic orientations in Sample 2. One hypothesis is that new nucleation of quartz occurred on the surface of existing quartz crystals during the formation of graphic granite. Therefore, they seem attached to each other. However, in such a case, the nuclei should have an epitaxial relationship with the host quartz crystals, and we infer that this hypothesis is not likely. The other hypothesis is that new nucleation and branch formation of quartz occurred independently of other quartz crystals, but they were attached to each other during the growth.

Although the reason for the difference in the nucleation of the two samples remains unclear, we can attribute such a difference to the supercritical state for example. The effect of the accumulated water on the genesis of pegmatites, that is, the decrease in the viscosity of the melt and increase in the diffusivity of the constituent elements, which enhance the formation of giant crystals, was strongly pointed out in the landmark paper of this field by Jahns and Burnham (1969). Researchers today fundamentally accept the role of aqueous fluid in the pegmatite formation, as described above. Jahns and Burnham (1969) also suggested that the aqueous fluid probably reaches a supercritical state in many cases during pegmatite formation, although it depends on the depth of the pegmatite body. One important role of supercritical water is to increase the solubility of materials and thus to enhance the transportation of materials, leading to the formation of giant crystals in pegmatites. On the other hand, the microscopic viewpoint, supercritical conditions cause a big fluctuation in the density (spatial distribution of water molecules). The degree of such a fluctuation in the supercritical water dramatically changes by a small difference in the pressure and temperature conditions (e.g., Nishikawa et al., 2004). It is also known that a slight change in the pressure and/or temperature near the critical point dramatically influences the nucleation and growth of the solute (e.g., Adschiri et al., 1992). The nucleation and growth of quartz rods with different crystallographic orientations in Sample 2 might be related to the change of the properties of supercritical water near the critical point.

Origin of the sharp boundary between the graphic and single-crystalline K-feldspar parts and a new model of pegmatite genesis

In Figure 8, we show the sharp interface between the graphic and single-phase parts of K-feldspar observed by X-ray CT. The most important finding of this study is that such a sharp interface was formed during the crystallization process of pegmatite. K-feldspar continued its crystallization as single crystals beyond this interface, but the growth of quartz crystals suddenly stopped at this interface. It is reasonable to believe that the physical or chemical conditions somewhat changed at this interface. Here, we discuss the formation process of pegmatites based on the sharp boundary between the graphic and single-crystalline K-feldspar parts observed by X-ray CT regarding the question whether the graphic granite resulted from eutectic or simultaneous crystallization of quartz and feldspar under non-equilibrium conditions. Barker (1970) denied the eutectic crystallization and described that the bulk chemical composition has no clear relation with any known eutectic point or coticetic curves between quartz and alkali feldspars. The chemical composition of the eutectic point in the water-saturated SiO2(Qz)-KAlSi3O8(Or) binary system is, for example, approximately 42(Qz):58(Or) (in weight percent) at 500 MPa (Johannes and Holtz, 1996), but the average proportion of the Qz component in the actual graphic granite is below 30 wt% (Barker, 1970), much smaller than that of the eutectic point. Prior crystallization of K-feldspar and the change of the melt composition to the eutectic point are necessary to cause eutectic crystallization. However, in actual graphic granite in pegmatites, K-feldspar coexists with quartz from the first stage of crystallization after

S. Ikeda, Y. Nakashima and T. Nakano

12
that from the Ishikawayama mine (e.g., Kondo, 1953; Jahns and Burnham, 1969; Mitsumori et al., 2000; London, 2009). Although this model assumes the intrusion of magma into the existing rock body with a lower temperature, it simply explains the zonal structure of pegmatites and the sharp interface between graphic granite and massive K-feldspar. Most of the natural pegmatites are intrusive and researchers have been considering recently that the pegmatites cooled quickly and a very large degree of undercooling occurs (e.g., London, 2005). In this sense, our model roughly agrees with the results of many previous studies. The novelty of our result is that the interface between graphic granite and massive K-feldspar corresponds to the transition point (surface) from non-equilibrium to equilibrium conditions; shortly after exceeding the boundary, the system reenters the ‘Kfs + Melt’ field in the KA\(_2\)Si\(_2\)O\(_6\)-SiO\(_2\) phase diagram from the subsolidus field. Further experiments, simulations, field work, and analyses of natural samples are necessary to verify the new model.

**CONCLUSION**

We investigated the boundary region between massive K-feldspar and quartz-feldspar intergrowth (graphic granite) using X-ray CT measurements and polarizing observations on thin sections. The biggest finding of this study is that the tips of the quartz rods create a sharp and flat interface between graphic granite and massive K-feldspar. The formation of such a structure can be explained by rapid cooling of the water-accumulated melt through the intrusion into a rock body with a lower temperature. After the emplacement of the magma, simultaneous crystallization of quartz and K-feldspar occurs, which is driven by large undercooling, although the chemical composition of the melt is away from the eutectic point (at the temperature below the solidus). With the growth of graphic granite, the equilibrium conditions recover due to the increase of the temperature and decrease of undercooling. At the same time, the growth front reenters the ‘Kfs + Melt’ field in the phase diagram, leading to the end of the quartz crystallization. This stage corresponds to the formation of the sharp interface between graphic granite and massive K-feldspar observed by X-ray CT. After stable crystallization of massive K-feldspar under equilibrium conditions in the ‘Kfs + Melt’ field, the chemical composition of the melt gradually changes toward the eutectic point (Fig. 9c), followed by the stable crystallization of quartz and K-feldspar under equilibrium conditions (Fig. 9d).

A zonal structure, such as that shown in Figure 9d, can be observed in many natural pegmatites including...
Figure 9. Proposed model showing the formation process of the zonal structure of pegmatites based on the results obtained in this study. (a) Formation of silicic magma with a high concentration of water accumulated during the crystallization of anhydrous minerals in a granitic pluton. (b) Intrusion of the magma into a lower-temperature rock body and formation of graphic granite driven by large undercooling (simultaneous crystallization of quartz and K-feldspar under non-equilibrium conditions). (c) Recovery of the temperature at the crystallization front and reentering the field of ‘Kfs + Melt’ in the phase diagram, leading to the growth of massive K-feldspar crystals under equilibrium conditions. With the crystallization of the massive K-feldspar, the chemical composition of the melt gradually changes toward the eutectic point. (d) Growth of massive crystals of quartz (gray) and K-feldspar (white) at the eutectic points under equilibrium conditions. The schematic phase diagram shown in this figure is roughly based on the phase diagram of the water-saturated SiO₂(Qz)-KAlSi₃O₈(Or) binary system, for example, approximately 42(Qz):58(Or) (in weight percent) at 500 MPa (Johannes and Holtz, 1996). In reality, the system contains a certain amount of an albite component and the temperature of the eutectic point decreases more (e.g., to ~650 °C), although the temperature of the eutectic point of the water-saturated Qz-Or system is 700–750 °C.
In this study, we also demonstrate the 3-D connectivity and single-crystalline degree of quartz crystals based on the cluster labeling analysis using X-ray CT data and polarizing observations using thin sections, respectively. The single-crystalline degree (simultaneous extinction) of quartz rods of the two samples differs, although the connectivity is high in both samples. Although uncertainty still remains regarding this problem, such differences can be explained with supercritical water; if the aqueous fluid in the system reaches a supercritical state, the probability of nucleation and formation of new crystals (with different crystallographic orientations) is largely changed by the slight change of the conditions (temperature and pressure), especially if the system is near the critical point of the supercritical state.

ACKNOWLEDGMENTS

We would like to sincerely thank the late Mr. Yoshimasa Yabe (president of the Mazak Stone Corporation, Ishikawa town, Fukushima Prefecture, Japan) who permitted us to survey the pegmatite mine and collect graphic granite samples. We also thank Prof. Masaaki Shimizu (University of Toyama) for providing useful information on the geology of pegmatites in Ishikawa town. Helpful and constructive comments by two anonymous reviewers and Prof. Masaki Enami who handled our manuscript as an editor greatly improved the manuscript. We are grateful for their careful review. This research was supported by Research Fellowships of the Japan Society for the Promotion of Science (JSPS) for Young Scientists (to S.I.), JSPS KAKENHI (Grant Numbers JP19J07812 and JP20J08525), a grant from the Agency of Industrial Science and Technology (to Y.N.), and Tohoku University Global COE Program Global Education and Research Center for Earth and Planetary Dynamics (Leader: Prof. Eiji Ohtani) promoted by MEXT, Japan.

REFERENCES

Adschiri, T., Kanazawa, K. and Arai, K. (1992) Rapid and Continuous Hydrothermal Synthesis of Boehmite Particles in Subcritical and Supercritical Water. Journal of the American Ceramic Society, 75, 2615-2618.

Baker, D.R. and Freda, C. (2001) Eutectic crystallization in the undercooled Orthoclase-Quartz-H2O system: experiments and simulations. European Journal of Mineralogy, 13, 453-466.

Barker, D.S. (1970) Compositions of granophyre, myrmekite, and graphic granite. Geological Society of America Bulletin, 81, 3339-3350.

Brown, M.A., Brown, M., Carlson, W.D. and Denison, C. (1999) Topology of syntectonic melt-flow in the deep crust: Interfaces from three-dimensional images of leucosome geometry in migmatites. American Mineralogist, 84, 1793-1818.

Denison, C. and Carlson, W.D. (1997) Three-dimensional quantitative textural analysis of metamorphic rocks using high resolution computed X-ray tomography: Part II. Application to natural samples. Journal of Metamorphic Geology, 15, 45-57.

Denison, C., Carlson, W.D. and Ketcham, A. (1997) Three-dimensional quantitative textural analysis of metamorphic rocks using high-resolution computed X-ray tomography: Part I. Methods and techniques. Journal of Metamorphic Geology, 15, 29-44.

Ebel, D.S. and Rivers, M.L. (2007) Meteorite 3-D synchrotron microtomography: Methods and applications. Meteoritics & Planetary Science, 42, 1627-1646.

Fenn, P.M. (1986) On the origin of graphic granite. American Mineralogist, 71, 325-330.

Fersmann, A.E. (1929) VIII. Die schriftstruktur der granitpegmatite und ihre entstefung. Zeitschrift fur Kristallographie, 69, 77-104.

Hounsfield, G.N. (1973) Computerized transverse axial scanning (tomography): Part I. Description of system. British Journal of Radiology, 46, 1016-1022.

Hubbell, J.H. and Seltzer, S.M. (1996) Tables of X-ray mass attenuation coefficients and mass energy-absorption coefficients from 1 keV to 20 MeV for elements Z = 1 to 92 and 48 additional substances of dosimetric interest, NIST, Physical Laboratory, Physical Reference Data. http://physics.nist.gov/PhysRefData/XrayMassCoef/cover.html.

Ikeda, S., Nakano, T. and Nakashima, Y. (2000) Three-dimensional study on the interconnection and shape of crystals in a graphic granite by X-ray CT and image analysis. Mineralogical Magazine, 64, 945-959.

Ikeda, S., Nakano, T., Tsuchiyama, A., Uesugi, K., Suzuki, Y., Nakamura, K., Nakashima, Y. and Yoshida, H. (2004) Nondestructive three-dimensional element-concentration mapping of a Cs-doped partially molten granite by X-ray computed tomography using synchrotron radiation. American Mineralogist, 89, 1304-1313.

Ikeda, S., Nakano, T., Tsuchiyama, A., Uesugi, K., Nakashima, Y., Nakamura, K., Yoshida, H. and Suzuki, Y. (2017) Three-dimensional study by synchrotron radiation computed tomography of melt distribution in samples doped to enhance contrast. Mineralogical Magazine, 81, 1203-1222.

Jalans, R.H. and Burnham, C.W. (1969) Experimental studies of pegmatite genesis: I. A model for derivation and crystallization of granitic pegmatites. Economic Geology, 64, 843-864.

Jerram, D.A. and Higgins, M.D. (2007) 3D analysis of rock texture: quantifying igneous microstructures. Elements 3, 239-245.

Johannes, W. and Holtz, F. (1996) Petrogenesis and experimental petrology of granitic rocks. pp. 335, Springer-Verlag Berlin Heidelberg.

Kawano, Y. and Ueda, Y. (1967) K-Ar dating on the igneous rocks in Japan (VI) - granitic rocks, summary -. The Journal of the Japanese Association of Mineralogists, Petrologists and Economic Geologists, 57, 177-187 (in Japanese with English abstract).

Ketcham, A. and Carlson, W.D. (2001) Acquisition, optimization and interpretation of X-ray computed tomographic imagery: application to the geosciences. Computational Geosciences, 27, 381-400.
Uesugi, K., Suzuki, Y., Yagi, N., Tsuchiyama, A. and Nakano, T. (2001) Development of high spatial resolution X-ray CT system at BL47XU in SPring-8. Nuclear Instruments and Methods in Physics Research Section A, 467–468, 853–856.

Wellington, S.L. and Vineger, H.J. (1987) X-ray computerized tomography. Journal of Petroleum Technology, 39, 885–898.