CATHODOLUMINESCENCE STUDY OF QUARTZ RECRYSTALLIZATION IN CONTACT-METAMORPHOSED ROCKS OF THE SHIMANTO SUPERGROUP, KANTO MOUNTAINS, JAPAN

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Abstract

Shimanto Supergroup sandstones and shales in the Kanto Mountains, Japan, have been contact metamorphosed to hornfels by intrusion of the Tokuwa Batholith. Garnet-biotite geothermometry shows that the metamorphic temperatures of samples collected at distances of ~1 m to 2600 m outward from the batholith range from about 700℃ to 400℃. Cathodoluminescence (CL) imaging of quartz from these samples allows the CL characteristics of quartz to be evaluated as a function of known metamorphic temperatures. Contact metamorphism is shown to generate two basic kinds of CL features, homogeneous bright CL and mottled CL, which are characteristic respectively of metamorphism at high and low temperatures. Other CL features, such as dark and bright CL lines may be superimposed on mottled CL texture. CL imaging provide a basis for understanding recrystallization of quartz that involves changes in trace-element composition and crystal defect structures under the influence of contact metamorphism.

Published studies indicate that most quartz in unmetamorphosed Shimanto Supergroup sandstones and shales was derived from volcanic and plutonic sources. Volcanic and plutonic quartz is characterized by distinctive CL textures such as fine-scale zoning. None of the quartz in metamorphosed Shimanto sediments that we investigated displays these CL textures, which indicates that original CL features were erased during an early dehydration stage of contact metamorphism. New quartz CL textures were subsequently acquired that reflect the conditions that prevailed during later stages of fluid infiltration at higher metamorphic temperatures.

Key Words: cathodoluminescence, contact metamorphism, quartz, recrystallization, Shimanto

INTRODUCTION

The Shimanto Supergroup in the Kanto Mountains, Japan, is part of an accreted terrane, located in the outer belt of the Japan Arc, which formed during late Cretaceous to Tertiary time. Sediments of the Kobotoke Group, a subdivision of the Shimanto Super-

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group, were thermally metamorphosed during the Tertiary by intrusion of granodiorite and tonalite of the Tokuwa Batholith (Shimizu, 1986). Kagami and Taniguchi (2003) used petrologic techniques and garnet-biotite geothermometry to map metamorphic zones in Kobotoke sediments and to determine the temperatures at which contact metamorphism took place within these zones.

Subsequently, one of the authors (Boggs) analyzed selected samples of Kobotoke quartz by using cathodoluminescence imaging to investigate CL characteristics of quartz as a function of metamorphic temperatures and fluid conditions. Cathodoluminescence (CL) imaging reveals textural details of quartz that are not visible by petrographic or other techniques. These CL features provide information about the thermal and fluid-migration history of metamorphic rocks, as well as providing a means of differentiating metamorphic quartz from quartz of other origins (e.g., plutonic, volcanic). The most common CL features of metamorphic quartz are mottled texture and homogeneous (low contrast) CL, which are related to temperature of metamorphism and the relative abundance of activator ions in metamorphic fluids (e.g., Boggs and Krinsley, 2006).

Our objective in this study was to investigate the CL textures of quartz subjected to contact metamorphism at known temperatures (estimated by geothermometry) in order to evaluate the survival of relict CL textures in Kobotoke quartz during metamorphism and to determine the characteristics of new CL textures generated at different metamorphic temperatures. Thus, both the effects that elevated metamorphic temperatures have on original CL characteristics of quartz and the kinds of new CL features generated during thermal metamorphism were examined.

STRATIGRAPHY AND PREVIOUS WORK

The Shimanto Supergroup in the Kanto Mountains is divided into the Ogochi (Cretaceous), Kobotoke (Cretaceous), and Sagamiko/Setogawa (Tertiary) groups (Sakai, 1987; Editing Committee of the Geological Map of Yamanashi Prefecture, 1970). Only the Kobotoke Group is of interest in this study. The Kobotoke Group is composed dominantly of turbidite sandstones, mudstones, and conglomerate, with some intercalations of limestone, chert, and basic volcanic rocks. Kobotoke Group rocks in the northeastern part of the area are largely coherent, whereas those in the southwestern part have been disrupted by extensive tectonic shearing (Ogawa *et al.,* 1988).

Kobotoke Group rocks have been contact metamorphosed by intrusion of the Kofu Plutonic Complex. That part of the complex that crops out along the western part of the study area (Fig. 1) is called the Tokuwa Granodiorite (Shimizu, 1986). Sedimentary rocks of the Kobotoke Group were transformed by contact metamorphism into biotite hornfels with small amounts of cordierite-biotite hornfels, muscovite-biotite hornfels, and cummingtonite-biotite hornfels (Shimizu, 1986). Metamorphism created characteristic mineral zones, which extend outward from the pluton contact (discussed below).

This study focuses on CL analysis of quartz in hornfels-facies rocks in the contact

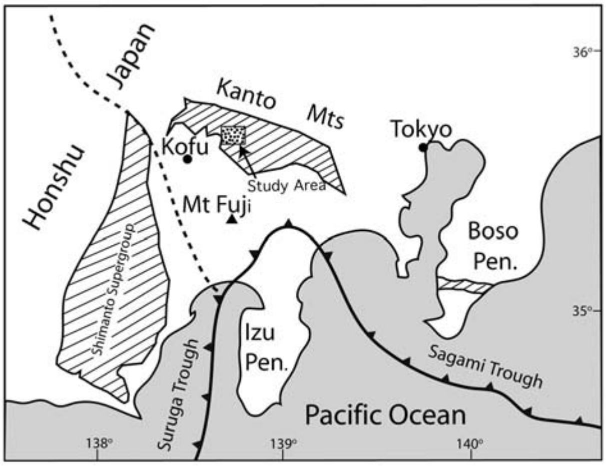

Fig. 1 Location map of the study area in the Kanto Mountains, central Honshu, Japan.

aureole of the Tokuwa Granodiorite. Previous investigation of these rocks includes an early field study of the hornfels in the aureole on the western side of the Kofu Basin initiated by Yuasa in 1976. Kagami and Taniguchi began field and laboratory study of metamorphic rocks in the Kanto Mountains in 1998 (Fig. 1) in the aureole to the east, outside the Kofu Basin, along tributaries of the Tama River. Stratigraphic studies of the Shimanto belt in the Kanto Mountains were carried out by the Editing Committee of the Geological Map of Yamanashi Prefecture (1970) and by Sakai (1987). Ogawa *et al.* (1988) did a detailed stratigraphic and structural review of the Shimanto Supergroup in the Kanto Mountains. Previous study of the CL characteristics of quartz in Kobotoke sediments has not been made.

METAMORPHIC ZONES AND SAMPLING SITES

Kagami and Taniguchi (2003) proposed contact metamorphic zones adjacent to the Tokuwa pluton along the tributaries of the Tama River. As shown in Figure 2, these zones extend from the garnet-cordierite zone, located along the contact, outward through the cordierite, biotite (1 and 2), and chlorite zones. A major focus of this study is evaluation of the cathodoluminescence characteristics of quartz as a function of contact metamorphic temperature. Therefore, samples were collected at five sites located at distances ranging from ~1 to 2600 meters from the pluton contact (Table 1). Metamorphic temperatures of the samples, which were taken from the garnet-cordierite and cordierite metamorphic zones, were estimated by garnet-biotite geothermometry to range from ~700℃ near the contact to ~400℃ outward from the contact (Kagami and Taniguchi, 2003; Kagami *et al.*, 2005).

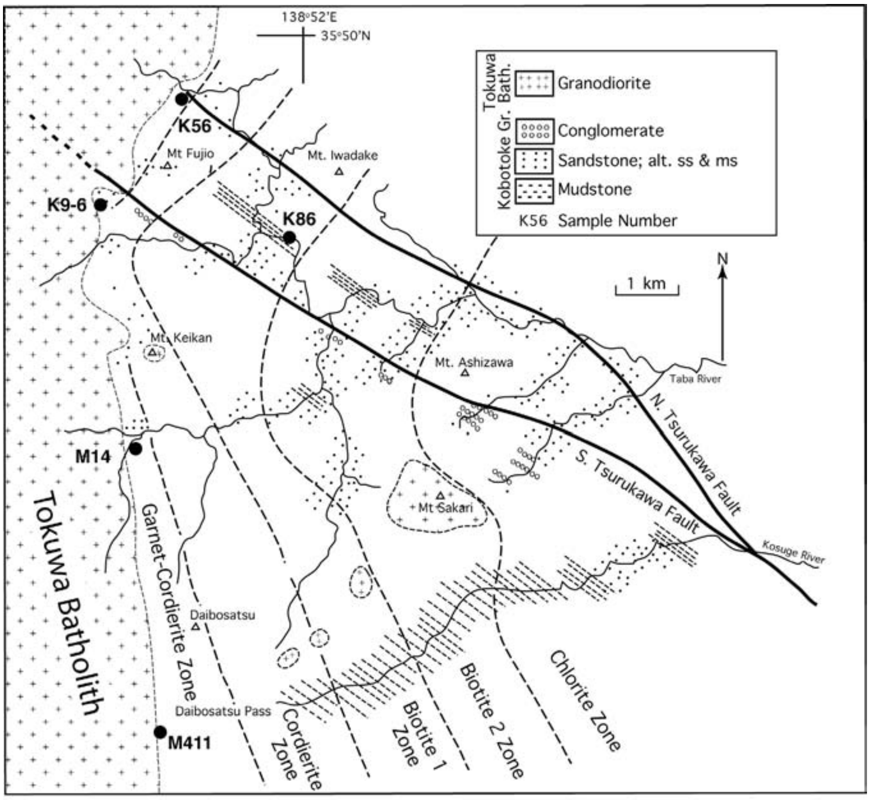

Fig. 2 Enlarged view of the study area showing generalized geology, metamorphic zones, and sampling sites.

Table 1 Characteristics of samples selected for cathodoluminescence imaging

Sample No.	Lithology	Dist. from Contact (meters)	Meta. Zone	Meta. Temperature (℃)
M411	Quartz segregation vein in sandy mudstone	1	Garnet-cordierite	600–680
K9-6	Sandstone	5	Garnet-cordierite	600–690
K56	Sandstone	50	Garnet-cordierite	650–710
M14	Mudstone & tuff	60	Garnet-cordierite	510
K86	Black Shale	2600	Cordierite	410–440

ANALYTICAL METHODS

Both standard petrographic microscopy and cathodoluminescence (CL) imaging were used to study each sample. Petrographic microscopy was carried out at the Ocean Research Institute, University of Tokyo. CL analyses of quartz were performed in the Department of Geosciences, University of Oregon by using a JSM-6300V scanning electron microscope (SEM) equipped with an Oxford Instrument mirror-type CL detector and a

Hamamatsu R374 photomultiplier tube. The SEM was commonly operated at 10 KV accelerating voltage, 5 nA beam current, and 22 mm working distance.

PETROGRAPHIC MICROSCOPY

Petrographic microscopy of selected samples shows that most Kobotoke sediments have a hornfels fabric consisting of nearly equidimensional crystals that display granoblastic to granoblastic-polygonal texture. An exception is Sample K86, which has a schistose fabric. Dominant minerals are biotite, quartz, and feldspars. Porphyroblasts of almandine garnet, cordierite, muscovite and oligoclase are also common. Very small veins of quartz (segregation veins) are present in some samples, e.g., samples K9-6 and M411. Most quartz grains in Sample K9-6 are characterized by irregular shapes and highly erose, embayed outlines. This sample also has an unusual mixture of coarse and fine grains of quartz and feldspar, which is a cataclasite fabric (Passchier and Trouw, 1996).

Selected textural features of quartz from the samples are shown in the Figure 3 photographs.

Sample K86. This plane-polarized image (Fig. 3-1) has a width of 0.2 mm with 20x objective. Quartz grains of anhedral shape are present in the schistose fabric. A quartz grain at the center of the photograph has a growth inclusion. Another grain at the left shows infiltration zoning at the periphery of the grain. Extinction of quartz is vague; overgrowths (dust rims) are rare.

Sample M14. Plane-polarized image (Fig. 3-2) has 0.2 mm width. Quartz grains are small equigranular tablets that form a polygonal fabric. Some grains display mosaic fabric, undulose extinction, and microcracks (Bernet and Bassett, 2005). A passive inclusion is present at the lower center of the photograph. Sharp extinction and nearly euhedral shape are remarkable features of the quartz grains at this site.

Sample K9-6. Plane-polarized image (Fig. 3-3), 0.2 mm width. The large grain at the center of the photograph shows fluid inclusion trails arranged in a belt-shaped distribution pattern across the grain. There is a healed fracture at the bottom of the photograph. Cross-polarized image (Fig. 3-4), 0.2 mm width. The quartz grains display interlobate and cataclasite fabric. Microfractures, infiltration zoning, and undulose extinction are common features.

Sample K56. Cross-polarized image (Fig. 3-5), 0.2 mm width. Coarse equigranular polygonal fabric is the dominant characteristics of this sample. The grain displays interlobate fabrics with microcracks near the center of the photograph. Mosaic fabrics, undulose extinction, growth inclusion , and fluid inclusion trails are additional features of grains at this site.

Sample M411. Plane-polarized image (Fig. 3-6), width 1.0 mm with 5x objective. This is the only example of vein quartz. Quartz crystals are coarse enough to be classified polygonal. Microcracks are present, but do not dislocate fluid inclusion trails. Sharp extinction is the striking characteristic of this quartz. Melt inclusions are common.

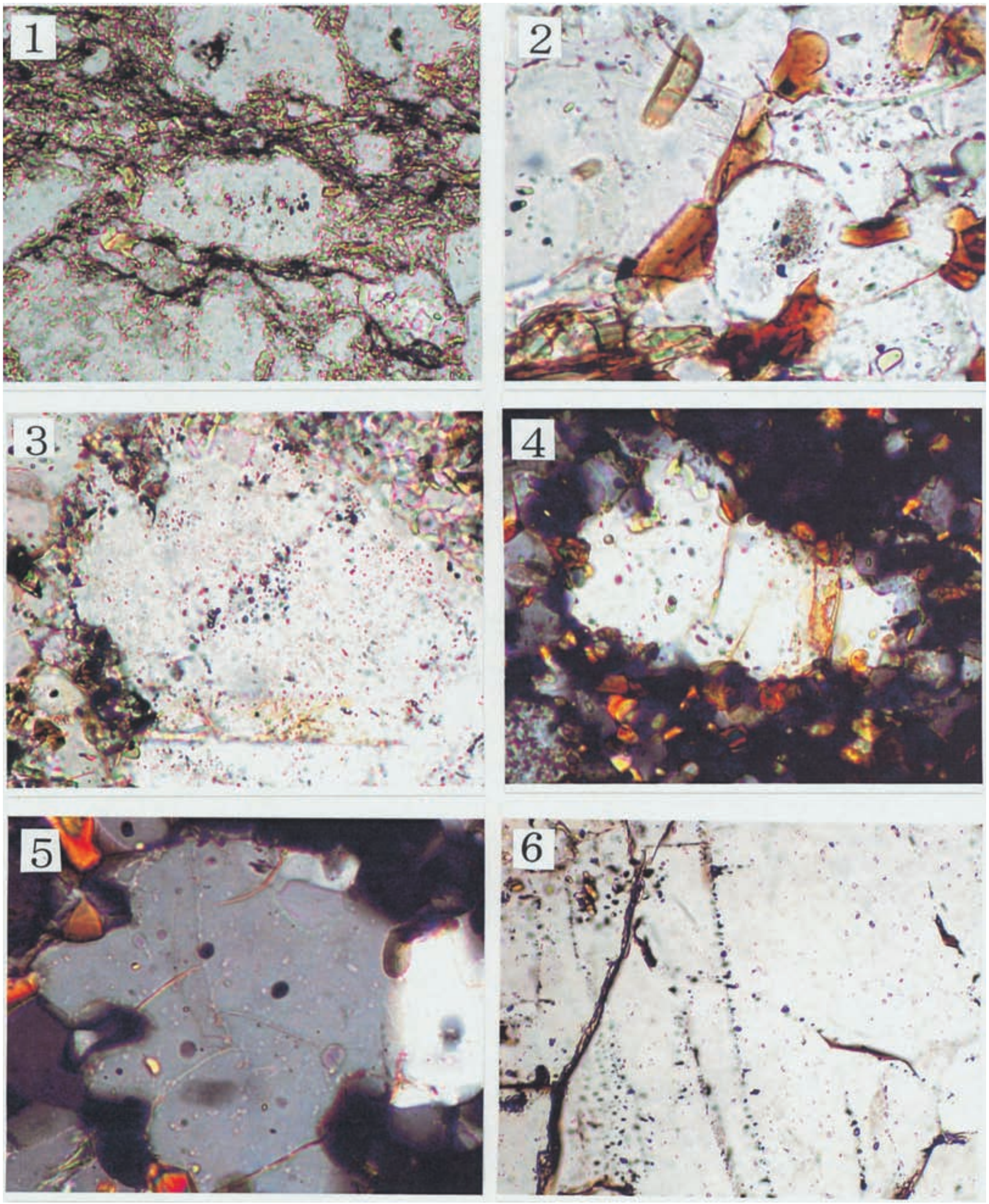

Fig. 3 Representative textural features of quartz crystal visible by petrographic microscopy

3-1 Growth inclusion in quartz grain, Sample K86 (Photograph No. 4255)
3-2 Passive inclusion in quartz grains, Sample M14-3 (No. 4221)
3-3 Fluid inclusion trails in quartz grains, Sample K9-6 (No. 4247)
3-4 Infiltration zoning, microfracture and interlobate fabric, Sample K9-6 (No. 4242)
3-5 Interlobate fabric and microcracks, Sample K56 (No. 4234)
3-6 Fluid inclusion trails and microfractues in quartz grains, Sample M411 (No. 39)

CATHODOLUMINESCENCE IMAGING

Cathodoluminescence imaging reveals textural details of quartz that arise from the presence of defects in quartz crystals that constitute electron traps. These defects may be either intrinsic (structural) defects or extrinsic defects caused by the presence of foreign ions (trace elements). When a crystal such as quartz is irradiated by a beam of high-voltage electrons, as in a SEM, electrons in the low-energy valence band of the crystal are promoted to a higher energy state in the conduction band. Electrons remain for only microseconds in the conduction band before losing energy and returning to the valence band. As de-energizing electrons move randomly through the crystal structure on their return path, they may be trapped momentarily by intrinsic or extrinsic defects. They then lose energy and vacate the trap; some of this lost energy is transferred to generation of photons, thus causing cathodoluminescence (e.g., Boggs *et al.,* 2001, 2002). The intensity of CL emission is related to the density of electron traps (defects) in the crystal; that is, increasing numbers of defects results in increasing CL emission (e.g., Marshall, 1988).

If defects are scattered more or less uniformly through a crystal, the crystal will display nearly uniform (homogeneous) CL. In many cases, however, defects are localized in certain parts of the crystal. Such a crystal will display nonuniform CL emission. That is, certain parts of the crystal will display bright CL and other parts dark CL. Thus, in general, the CL features of quartz crystals may include homogeneous CL, mottled CL, dark (black) CL streaks and patches, distinct CL zoning, and dark or bright CL lines (e.g., Seyedolali *et al,* 1997; Boggs and Krinsley, 2006). These CL features reflect the history of the quartz grain, e.g., crystallization from a plutonic or volcanic magma, metamorphism, tectonic deformation.

CL analysis of numerous metamorphic quartz grains (Boggs *et al.,* 2002; Boggs and Krinsley, 2006) shows that metamorphic quartz displays two main CL characteristics: homogeneous (uniform intensity) CL and mottled CL. These textural patterns are related to temperature of metamorphism and the presence of metamorphic fluids, to be discussed. Metamorphic quartz may also display other CL features such as distinct dark or bright CL lines, which commonly indicate fractures, and bright CL spots.

CL CHARACTERISTICS OF KOBOTOKE GROUP QUARTZ

Sample M411 (~1 m from contact) 600–680℃.

This sample was taken from a small segregation vein in sandy mudstone located very near the contact with the pluton. It displays nearly homogeneous CL with a slightly granular texture that resembles the texture of sharkskin (Fig. 4A). Other than slight, pen-point variations in CL intensity, the quartz grains display few other CL features. Note that the backscatter image (Fig. 4B) is also nearly featureless. The edges of many of the quartz grains are very irregular and are indented by embayments. The backscatter image

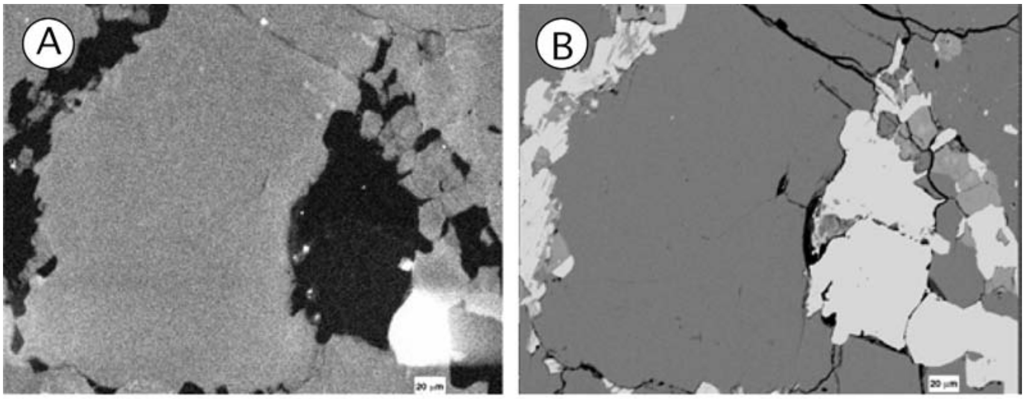

Fig. 4 (A) Cathodoluminescence image of quartz in Sample M411. Note that the quartz displays slightly granular, nearly homogeneous bright CL. (B) Backscattered SEM image of the same grains. The dark-CL areas in Fig. 4A indicate other minerals (biotite and feldspar), as shown in Fig. 4B.

shows that other minerals (mainly biotite and feldspar) fill the embayments. Also, very small mineral inclusions are present within some quartz grains. Open fractures are visible in backscatter images. On the whole, however, the outstanding CL feature of the quartz is nearly homogeneous CL.

Sample K9-6 (5 m from contact) 600–690℃.

Quartz in this sample displays mottled CL (less homogeneous than Sample M411). Grain boundaries tend to be highly irregular, which gives the grains an amoeba-like shape (Fig. 5) that is probably related to growth of adjacent biotite and feldspar crystals. The shape may also have been affected by grain-boundary migration related to shearing stresses arising from intrusion into Kobotoke sediments of innumerable granodiorite dikes (Kagami and Taniguchi, 2003). A few quartz grains contain dark CL lines, interpreted as fractures. SEM backscatter images show that these fractures are open; that is, they have

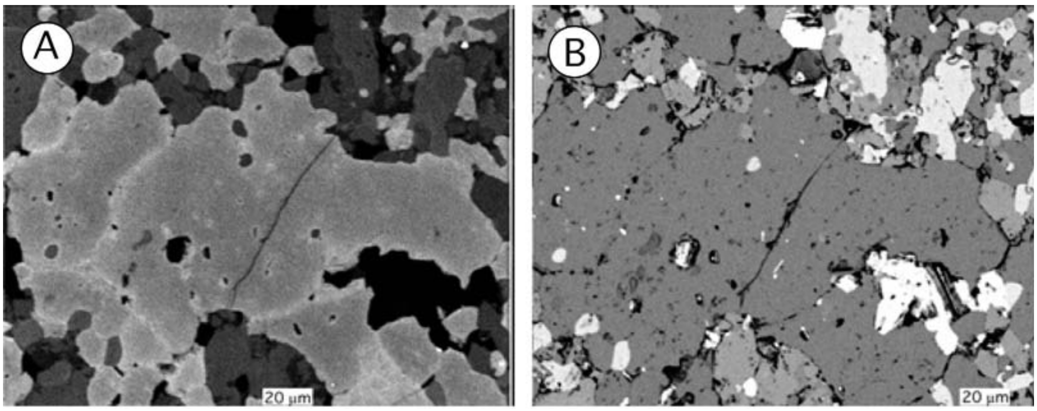

Fig. 5 (A) CL image of corroded and embayed quartz (Sample K9-6) that displays slightly mottled CL. The backscattered image (B) reveals that biotite and feldspars are present in the embayments. Dark spots in Fig. 5A are small mineral inclusions.

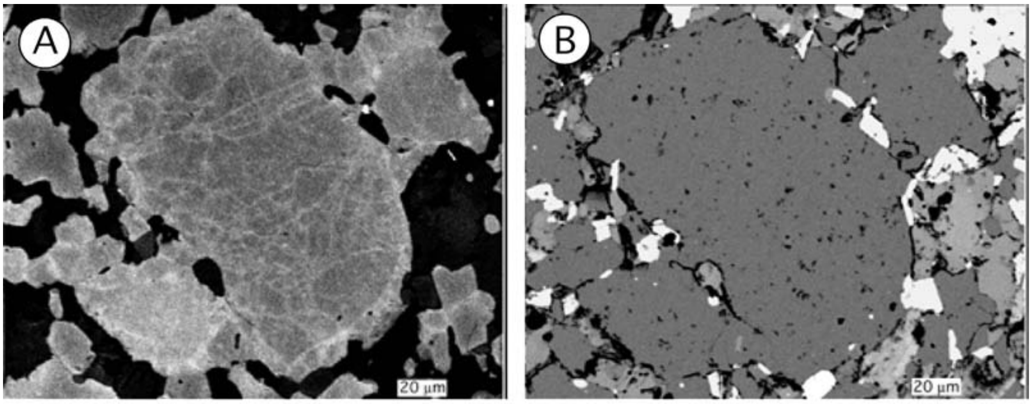

Fig. 6 (A) Bright CL "fracture" lines in Sample K9-6 quartz. The backscatter image (B) does not show these bright CL lines, indicating that the "fractures" are filled with quartz.

not been healed by subsequent precipitation of silica. Rare quartz grains exhibit a network of narrow lines that display bright CL (e.g., Fig. 6A). Small, dark CL areas within Fig. 6A are mineral inclusions.

Sample K-56 (50 m from contact) 650-710℃.

The quartz grains in this sample display some unusual CL features. Some grains exhibit small bright CL "spots" scattered through the grain (Fig. 7). These spots are not mineral inclusions, as shown by the backscatter image. They appear to be associated with isolated point lattice defects in the crystal. A few quartz grains are characterized by a network of bright CL lines, similar to that in Sample K9-6. Other grains exhibit a bright CL zone around the perimeter of the grain (e.g., Fig. 8). The contact between this bright CL zone and the interior of the grain is diffuse and gradational. Tiny mineral inclusions are present in this border zone. Many grains contain open fractures (dark CL lines), and

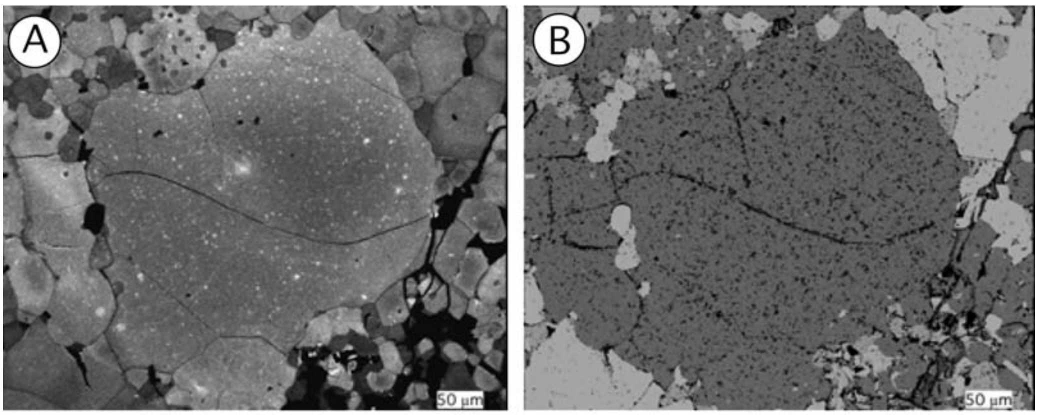

Fig. 7 (A) Cl image of quartz in Sample K56 showing bright CL spots that may represent point structural defects. Note the open fractures (dark CL lines). (B) Backscatter image of the same grain.

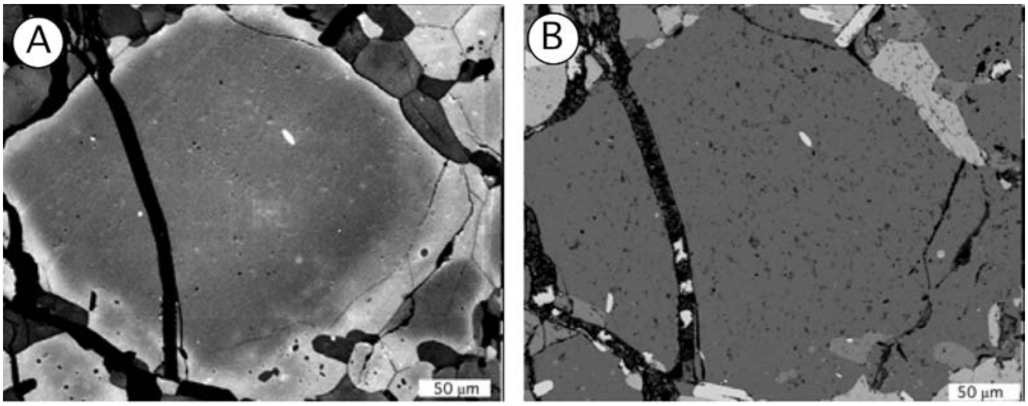

Fig. 8 (A) A Sample K56 quartz grain that displays a bright CL rim, which grades gradually to the darker CL grain interior. (B) Backscatter image of the same grain.

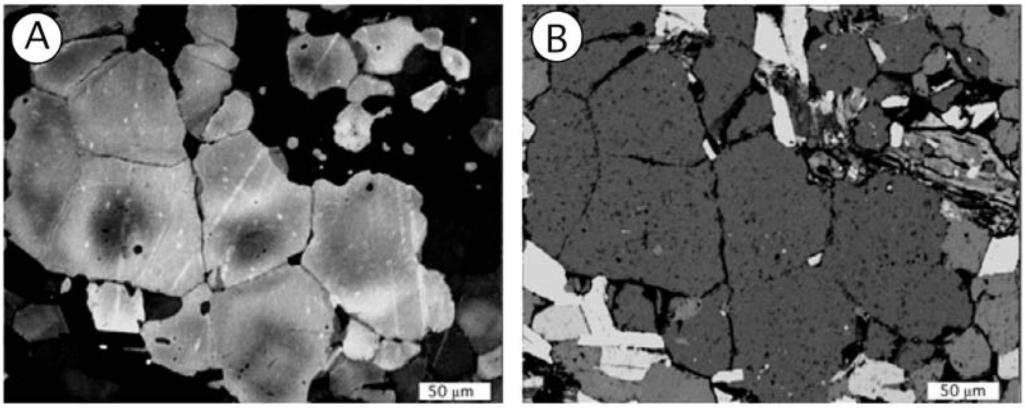

Fig. 9 (A) CL image showing distinctly mottled CL texture and fractures in Sample M14. Note that most of the fractures appear as distinct black lines in the backscatter image (B), indicating that they are open fractures.

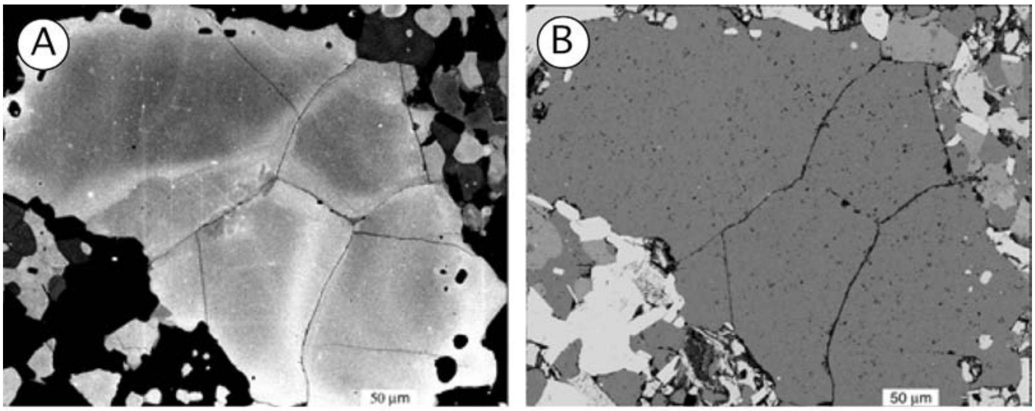

Fig. 10 (A) A Sample K86 quartz grain that displays mottled CL, open fractures, and gradational zones of bright CL around the perimeter of the grain and along open fractures. (B) Backscatter image of the same grain.

rare grains display bright CL lines.

Sample M14 (60 m from contact) 510℃.

The CL of quartz grains in this sample is similar to that of Sample K–56; however, some of the quartz displays a more distinct mottled texture (Fig. 9). The peripheral zone of bright CL in some grains broadens to encompass much of the interior of the grains. Tiny spots of bright CL are present in some grains. Open fractures are moderately common; healed fractures (fracture lines not visible in backscatter images) are relatively uncommon.

Sample K86 (2600 m from contact) 410–440℃.

This sample is located within the cordierite metamorphic zone more than two kilometers from the pluton contact. The metamorphic temperature is the lowest of any site. Most quartz grains display well developed mottled CL texture. Bright CL zones with gradational boundaries are located along the perimeters of many grains, as well as along many fractures (Fig. 10). Backscatter images indicate that many of the fractures are open fracture. Irregular grain boundaries that contain embayments filled with biotite and feldspar are common. Small mineral inclusions are present in the bright CL rim zones of some grains.

SUMMARY OF QUARTZ CL FEATURES

Two primary, end-member, types of quartz are recognized in Kobotoke Group samples on the basis of CL textures: quartz characterized by nearly homogeneous, moderately bright CL texture and that characterized by mottled texture. Homogeneous, moderately bright CL texture is present in quartz mainly from sites near the pluton contact, particularly quartz in segregation veins. Quartz characterized by bright, homogeneous CL displays few other CL features. Mottled texture is characteristic of quartz metamorphosed at somewhat lower temperatures at sites located greater distances from the contact. Gradations exist between these end-member types, that is, quartz CL that is almost but not completely homogeneous such as in Sample K9-6 located 5 m from the contact.

Several other kinds of CL features may be superimposed on mottled or slightly mottled CL texture. Bright CL edge zones are present along the periphery of some grains, as well as along open fractures (dark CL lines). These bright zones grade imperceptibly to darker CL in the grain interiors. Open fractures, which cut across all types of CL features, are present on most grains; however, healed fractures (filled with precipitated dark CL silica) are relatively uncommon. Bright CL lines and small bright CL spots are present on rare quartz grains.

INTERPRETATION OF CATHODOLUMINESCENCE

Homogeneous CL.

Quartz in Sample M411 is interpreted to have originated as a precipitate from hot fluids within a segregation vein near the pluton contact. Silica may have been furnished by reaction with adjacent wall rock (e.g., Barker, 1998, p. 181). The quartz displays bright, nearly homogeneous but slightly granular CL texture (Fig. 4). Nearly homogeneous bright CL indicates high density of luminescence centers (electron traps), which are spread more or less uniformly throughout the quartz crystal. Homogeneous bright CL is not a common primary CL texture in quartz; its presence in quartz from the segregation vein suggests that original CL texture (probably zoning) was erased by subsequent metamorphism. High luminosity indicates that additional luminescence centers formed during metamorphism owing to diffusion of CL-activating trace elements into quartz to create extrinsic defects. These processes are considered further in subsequent discussion.

Mottled CL.

Mottled CL texture (e.g., Fig. 9A) indicates irregular distribution of activator ions or defect structures within quartz grains (e.g., Boggs and Krinsley, 2006). That is, brighter CL areas within quartz grains contain more activator ions or defect structures than do darker areas. The exact mechanism by which mottled CL texture forms is poorly understood. It appears to develop at metamorphic temperatures high enough to erase relict textures such as zoning but not high enough to generate homogeneous bright CL. After relict textures are removed, CL-activating trace elements in surrounding fluids apparently diffuse irregularly into the crystal to produce the mottling.

It is possible that some quartz grains in Kobotoke sediments were derived from metamorphic source rocks and may still retain original (relict) mottled CL texture. That is, the mottled texture may have been inherited from a previous episode of metamorphism. On the other hand, Sakai (1987) reports, on the basis of petrographic studies of Shimanto Supergroup sediments, that sandy sediment in Kobotoke rocks in this area consist mainly of materials derived from felsic to intermediate volcanic rocks with only minor amounts of grains derived from granitic plutonic, sedimentary, and metamorphic source rocks. Volcanic and plutonic quartz grains are characterized by distinctive CL textures (e.g., fine-scale zoning). Given that we have not observed relict volcanic or plutonic CL textures in metamorphosed Kobotoke quartz, it appears unlikely that much Kobotoke mottled-CL quartz is truly relict. Most of the mottled CL was produced during contact metamorphism of Kobotoke Group sediments. Generation of mottled CL in quartz is also considered further in a succeeding discussion section.

Bright CL Zones along Grain Margins and Open Fractures.

As shown in Figure 8, bright CL edge zones are present around the periphery of some grains. These bright zones display gradual gradation to darker CL interiors of the grains. Holness and Watt (2001) reported fine-scale alternating bands of bright and dark CL around grain margins of contact-metamorphosed quartz, which they attributed to grain growth during metamorphism. Kobotoke quartz grains do not display such sharp planar boundaries. Gradual gradation of bright CL to darker CL indicates, instead, that these zones form by alteration, not grain growth. Trace-element rich metamorphic fluids that were present around grain margins and within fractures in grains reacted to introduce CL-activator ions (trace elements) into the grains in interstitial or substitutional positions in the quartz lattice (e.g., Götze *et al.,* 2004). The presence of these CL bright zones indicates that fluids played an important role in the metamorphic process.

Bright CL Spots.

Tiny spots of bright CL that are widely distributed through some quartz grains in Sample K56 (Fig. 7) and a few other samples are unusual and difficult to explain. Their small size suggests that they represent scattered point defects of some kind. Barker (1998) reports that point defects in metamorphic crystals can result from vacant sites in the crystal lattice or from extra atoms or molecules in interstitial or substitutional positions. Apparently, point defects may migrate through the crystal lattice by diffusive processes involving exchange with neighboring ions. The specific metamorphic process that was responsible for creating the rare point defects in Kobotoke Group quartz is not known. The bright CL spots are most common in quartz located ~50–60 m from the pluton contact.

Dark CL Lines.

Thin, dark CL lines indicate the presence of fractures (e.g., Seyedolali *et al.,* 1997). Fractures that appear in both CL and backscatter images are open fractures; fractures that appear only in CL images are healed fractures, which have been filled with SiO_2 precipitated at low temperature. Healed fractures are comparatively rare in Kobotoke quartz. On the other hand, open fractures are very common. Many of these fractures cut across other CL textures, which indicates that they are late-stage features that were probably generated by tectonism.

Bright CL Lines.

Rare Kobotoke quartz grains contain a network of bright CL lines rather than dark CL lines (e.g., Fig. 6). These bright CL lines are not visible in backscatter images, and they are very faint in CL images in comparison to dark CL fracture lines. They appear to be incipient fractures that were healed by precipitation of silica at moderately high temperatures or by recrystallization at high temperature accompanying infiltration by fluids. Holness and Watt (2001) observed similar features in the Appin Quartzite and proposed a similar interpretation. Because the grains that display networks of these bright CL lines are so rare, they may be an inherited relict feature.

DISCUSSION

Homogeneous CL.

Homogeneous CL is a characteristic CL texture of much metamorphic quartz (Seyedolali *et al.,* 1997; Boggs and Krinsley, 2006). Homogeneous CL with dull luminescence (dark CL) can be caused by nearly complete removal of CL-activating defects from the crystal structure, as suggested by Matter and Ramseyer (1985). Matter and Ramseyer state that loss of cathodoluminescence reflects cleaning of the quartz crystal structure from trace elements and structural defects, but they provide little discussion of how this cleansing takes place. Presumably, CL-activating trace elements are mobilized at higher temperatures and removed entirely from the crystal by outward diffusion. Also, structural defects are eliminated (healed or annealed) at moderate metamorphic temperatures. For example, Barker (1998, p. 131) states that elimination of dislocations and other lattice defects at elevated temperatures is an integral part of the process of static recrystallization. These processes likely occurred in Kobotoke sediments during an early dehydration stage of prograde metamorphism when fluids were released from hydrous minerals, as discussed by Bucher and Frey (2002). Thus, thorough cleansing of the crystal lattice of both trace elements and structural defects removes original CL textures (such as the zoning common in volcanic quartz) and generates homogeneous, low-intensity (dark) CL.

Sprunt, Dengler, and Sloan (1978), report that CL color of metamorphic quartz is related to metamorphic grade. They state that metamorphism appears to homogenize luminescence and that low temperature causes red luminescence and high temperature, blue. This observation is consistent with that of Boggs *et al.* (2002), who noted that low-temperature metamorphic grains display red CL and tend to have very low CL luminosity (dark CL) whereas high-temperature metamorphic grains display blue CL and have high overall luminosity (bright CL).

Homogeneous dark CL of thermally metamorphosed quartz can apparently be generated during a single stage of thermal metamorphism during which the quartz crystal lattice is cleansed of most CL activators. On the other hand, generation of homogeneous bright CL in quartz, such as that of Sample M411 quartz (from a segregation vein), requires an additional stage of higher temperature metamorphism during which trace elements are mobilized and diffused into quartz in substantial numbers to create extrinsic (trace-element) defects. Quartz in Sample M411 precipitated from hot, silica-rich fluids in a segregation vein (e.g., Walther and Wood, 1986), which may have acquired silica by diffusion from the matrix of Kobotoke sandy mudstone. Quartz was likely characterized initially by CL growth zoning, similar to that typical of hydrothermal quartz (e.g., Rusk and Reed, 2002). No trace of zoning now remains. Absence of relict CL zoning suggests that Sample M411 quartz was subjected (after initial formation) to continuing metamorphism that homogenized CL, as reported by Sprunt, Dengler, and Sloan (1978)). Subsequent metamorphism in the presence of fluids caused diffusion of additional CL-activating trace elements into the quartz to increase CL brightness. Several trace elements are recognized to be important CL-activators in quartz. For example, Al^{3+} and Ti^{4+} are characteristic impurity ion that substitutes for Si^{4+} in the silicon-oxygen tetrahedra, whereas Li^{+}, Na^{+}, K^{+}, Fe^{2+}, and H^{+} are ion compensators that enter interstitial positions. (e.g., Götze *et al.,* 2004; Müller, Seltmann, and Behr, 2000).

Diffusion of trace elements into quartz requires the availability of metamorphic pore fluids rich in these elements. The presence on many quartz grains of bright CL rims that grade to dark CL interiors, such as in Figure 8, provides evidence that such fluids were present. The highly embayed grain boundaries in Figure 5 are further evidence of the common presence of metamorphic fluids. In general, metamorphic fluids exert an extremely significant influence on mineral reactions during metamorphism (e.g., Walther and Wood, 1986).

Mottled CL Texture.

Quartz from sample sites located more distant from the pluton contact (and at lower temperatures) is distinguished by mottled CL texture, a feature characteristic of many quartz grains derived from metamorphic source rocks (Seyedolali *et al.,* 1997; Holness and Watt, 2001). Some mottled CL texture in Kobotoke quartz could have been inherited from a metamorphic precursor; however, as mentioned, Sakai (1987) reported that Kobotoke sandstones in the Kanto Mountains were derived from volcanic and, to a much lesser extent, plutonic sources. We have not observed quartz with relict volcanic (or plutonic) CL features in Kobotoke Group sediments that have undergone contact metamorphism; therefore, volcanic and plutonic CL textures, such as fine-scale zoning, must have been erased by contact metamorphism and replaced by mottled or homogeneous CL textures that were generated by contact metamorphism.

Little published information is available that lends insight into the process by which metamorphism of quartz generates mottled CL texture. Presumably, it could form by a one-step process that simply involves incomplete (patchy) "cleansing" of the crystal structure. In that case, distinctive relict CL features, such as of fine-scale zoning, should remain in those parts of the grain not thoroughly cleansed of activators. So far, we have not observed such relict traces in metamorphosed Kobotoke quartz, which indicates that the generation of mottled CL is a more complex process.

The process presumably begins by thorough cleansing of the crystal lattice to produce nearly homogeneous dark CL, as described above, during an early dehydration stage of prograde metamorphism when hydrous minerals such as chlorite and biotite release water owing to dehydration. Field evidence for an early dehydration stage of fluid generation is provided by mineral assemblages in the chlorite, biotite, and cordierite metamorphic zones (Kagami *et al.,* 2005). In a subsequent stage of metamorphism, called infiltration metasomatism (e.g., Bucher and Frey, 2002), trace elements diffused irregularly ("patchily") into the crystal lattice to create areas of brighter CL within darker CL areas. During this stage, sheet silicates and other hydrous minerals were consumed, and the cordierite —

K-feldspar and cordierite-garnet subzones were created. This high-temperature stage of metamorphism is associated with fluids containing abundant dissolved elements, including CL-activating trace elements such as Al^{3+} and Fe^{2+}. Evidence for infiltration metasomatism is provided by whole-rock chemical analyses, which indicate that element-rich fluids were commonly present during late-stage metamorphism, particularly near the contact with the pluton (Kagami *et al.,* 2005).

Invasion of trace-element rich fluids along incipient or open fractures (e.g., Fig. 5 and 10) and along grain margins may account for much of the observed mottling. Although mottled CL texture is particularly common in low-temperature metamorphic (greenschist facies) quartz, it has also been observed in higher-grade (i.e., amphibolite facies) quartz (e.g., Boggs and Krinsley, 2006). Sample K9–6 (Fig. 5) is an example of higher grade, CL-mottled quartz formed in Kobotoke sediments by dynamic recrystallization.

SUMMARY AND CONCLUSIONS

Contact metamorphism of Kobotoke Group (Shimanto Supergroup) sediments by intrusion of the Tokuwu Batholith generated metamorphic temperatures ranging from nearly 700℃ at the pluton contact to about 400℃ at a distance of 2600 m. Under the influence of metamorphic temperatures and fluids, quartz was affected by processes that included annealing of crystal defects and gain and loss from the crystal structure of CL-activating trace elements. Cathodoluminescence imaging shows that original CL textures (e.g., fine-scale zoning) in quartz near the pluton contact were erased and replaced by nearly homogeneous, bright-CL texture. Contact metamorphism of quartz at greater distances from the contact, and at lower temperatures, generated mottled CL texture.

Generation of homogeneous- and mottled-CL textures appears to be a two-stage process that involves reaction of quartz grains with metamorphic fluids. First, CL-activating trace elements diffuse out of quartz grains in the presence of fluids generated by dehydration reactions (accompanied by healing of structural defects), which erases relict CL textures and produces homogeneous dark CL. Pore fluids during this stage of metamorphism are apparently depleted in trace elements. During a subsequent infiltration metasomatism stage, trace-element rich pore fluids provide activator ions that diffuse into the quartz grains, as suggested by Götze *et al.* (2004), to generate brighter CL. Homogeneous, bright CL is produced at high metamorphic temperatures, which enhance the mobility of ions and allow trace elements to diffuse uniformly throughout the quartz crystal. At somewhat lower temperatures, ion mobility is retarded and trace elements diffuse patchily into parts of the crystal (e.g., along fractures) to generate mottled CL.

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