Origin of iridescence in grandite garnet

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Abstract

Iridescence in grandite garnet from Kamihogi, Yamaguchi Prefecture, Japan, results from two chemically distinct kinds of growth lamellae, which repeat alternately parallel to (110) with a periodicity of about 2 μ m. Thin sections of grandite garnets from Adelaide mining district, Nevada, show both straight and wavy lamellae with chemical differences. The internal textures correlate with growth features of high-index crystal surfaces. The internal texture was produced during growth, not by exsolution. Iridescence results from fine chemical lamellae repeating at a constant interval of less than about 6 μ m, not from twinning.

Introduction

Iridescent garnet was first collected at Adelaide mining district, Nevada, in 1934 (Barksdale, pers. comm., 1982), and was described by Ingerson and Barksdale in 1943. Fine lamellae optically observed in this garnet are oriented parallel to the (110) and (111) faces. In a section that was heated almost to the melting point (1225°C), the birefringence was very low, but growth bands which looked like polysynthetic twinning and iridescence were still visible. It appeared that the iridescence was due to the very fine (111) lamellae and that it was more intense where individual lamellae were finer.

Hirai and Nakazawa (1982) studied Adelaide iridescent garnet with the chemical composition $And_{90}Gr_{10}$ and suggested that iridescence results from periodic twins parallel to the (110) growth layers with a periodicity of about 1000Å, because an analytical transmission electron microscope did not detect any compositional difference across the boundaries of the lamellae. However, they did not show any evidence of twinning or suggest a formation mechanism for the twins. Hirai et al. (1982) suggested that all optical lamellae observed in thin sections of Adelaide garnet were produced after crystal growth, because the wavy lamellae cross the (110) growth zoning and the (110) straight lamellae are joined to the wavy lamellae.

Akizuki (1984) studied the relations among growth sectors, optical anomalies, and Fe^{3+}/Al ordering of grandite garnets from several localities including Kamihogi, Japan, where iridescent grandite garnet was found, and suggested that the optically non-cubic Fe^{3+}/Al ordered structure of garnet was produced during growth.

The objective of the present study is to describe the texture of iridescent garnets from Kamihogi, Japan, and Adelaide mining district, Nevada, and discuss the origin of the iridescence.

Specimens

Garnet from Kamihogi, Yamaguchi Prefecture, Japan, occurs in contact metasomatized limestone as aggregates of greenish-brown translucent crystals with well-developed {110} faces about 1 cm across. The average chemical composition is $And_{82.9}Gr_{11.3}Py_{5.5}Sp_{0.3}$ (Ito, 1940). Although Kamihogi garnet specimens were collected several decades ago, specimens are no longer found at that locality (see Sadanaga and Bunno, 1974). Surface features on the (110) faces and internal sectoral textures of Kamihogi garnet were described by Akizuki (1984).

Adelaide iridescent garnets were kindly provided by Prof. E. G. Ehlers through Prof. J. D. Barksdale. The garnets consist of small (110) and large (hh.2h) and (hk0)and (hkl) faces such as (112), (118), (120), (510), and approximately (90.11.90), on which striations parallel to (110) are commonly observed. It is difficult to determine the Miller indices of these faces, because of striations. Crystal faces with high indices are not common in nature, though such faces have been summarized by Dana (1920). The Adelaide specimen was described in detail by Ingerson and Barksdale (1943).



Fig. 1. Optical micrograph of areas of fine lamellae showing iridescence (light color) observed in a thin section normal to a (110) face. The lamellae are normal to the section. Kamihogi garnet.

Observations

Kamihogi garnet

The crystals consist of well-developed {110} sectors with small {112} sectors, though (112) faces are not found



Fig. 2. Fine growth lamellae parallel to a (110) face (SEM back-scattered electron image) and corresponding X-ray AlK α line profile. Aluminum content is greater to the left in the line profile. The thin section is normal to the (110) face. Kamihogi garnet.

on the surface. Figure 1 exhibits an optical photomicrograph of fine lamellae found in thin sections cut normal to the (110) growth surface of the garnet. The light areas, which consist of fine lamellae parallel to the growth surface, show iridescence. Iridescence was observed in this thin section normal to the (110) growth plane with a tilting stage in both transmitted and reflected light, whereas iridescence was not observed in thin sections cut parallel to (110). The color varies with change in direction of the incident light beam and also varies in different parts of the crystal. Some lamellae consist of colored domains. Although the garnet crystals from Kamihogi were found several decades ago, the iridescence was not noticed until the present study. Distinct birefringence is observed in the crystal between crossed polars (Akizuki, 1984).

Crystals polished normal to the growth plane (110) were studied with an electron microprobe to ascertain the chemical differences across the lamellae. Aluminum content increases as iron decreases, a relationship that is generally observed in grandite garnets. Figure 2 shows a compositional image and corresponding Al $K\alpha$ line profile. These figures indicate that the black lamellae are higher in aluminum than the white lamellae. Aluminum-rich lamellae typically are thinner than iron-rich lamellae, though this relationship is reversed in some areas. The



Fig. 3. Fine growth lamellae (SEM back-scattered electron image). The thin section is normal to the (110) faces. See text for details. Kamihogi garnet.



Fig. 4. Surface texture with triangular terraces on a (201) face, observed with SEM. See text for details. Adelaide garnet.

wavelength of the lamellae varies from several microns to several thousand ångströms in different parts of the crystal. The periodicity is slightly variable even in a small area (Fig. 2), and therefore the iridescence over most of the crystal is weak, though it is very strong in some lamellae with regular lamellar spacing. The crystal consists of many small growth sectors (Akizuki, 1984), and the lamellae kink at the sector boundaries but extend



Fig. 5. Surface texture consisting of grooves and fine growth steps on the same surface shown in Fig. 4. Vertical broad grooves transform into fine horizontal grooves at the bottom. SEM. Adelaide garnet.



Fig. 6. Wavy lamellae in thin section cut parallel to a (501) face, on which corresponding wavy grooves exist. SEM back-scattered electron image. Adelaide garnet.

continuously through them (Fig. 3). The lamellae labeled D in Figure 3 are the finest observed in the specimen, with a distance between two adjacent black lamellae of about 8000Å. The spacings of lamellae A and B in Figure 3 are similar, while lamellae D are thinner than lamellae C, because of a difference in growth rate. The areas with lamellae (A, B, C, and D) all show iridescence.

Adelaide garnet

Although Hirai et al. (1982) described the internal textures of Adelaide garnet, the present work adds some important observations. Surface features were observed by means of scanning electron microscopy (SEM) and reflection-type interference contrast microscopy. Figure 4 shows triangular terraces on a (201) growth surface. The outlines of the terraces presumably are parallel to the rhombohedral {110} faces. Also, Figure 4 shows horizontal straight striations parallel to (110) and wavy lamellae crossing the striations; each triangular terrace is composed of smaller triangular terraces. The wavy lamellae change continuously to broad grooves without the terraces crossing the striations on the same surface (Fig. 5). The orientations of the grooves vary abruptly or gradually with variation in inclination of the surface, and the broad grooves transform into fine grooves, which are inclined slightly to the striations (Fig. 5). Although some naturally etched garnets from other localities show characteristic small etch pits on the (110) face, no evidence of natural etching was found in the present specimen, suggesting that the surface features are a growth pattern.

After the growth surface (501) with the wavy grooves was slightly polished, a thin section parallel to this face was prepared. Wavy lamellae corresponding to the wavy



Fig. 7. Horizontal fine straight lamellae and inclined broad wavy lamellae. SEM back-scattered electron image. Adelaide garnet.

grooves on the surface were observed with a polarizing optical microscope, electron microprobe, and SEM (Fig. 6). Lamellae with high Fe concentration (light color in Fig. 6) are optically isotropic or nearly isotropic, while lamellae with a relatively higher concentration of Al (dark color), which correspond to the grooves on the surface, are anisotropic, and their extinction angles are inclined about 35° to (010) on the (501) plane.



Fig. 8. Wavy lamellae and horizontal straight lamellae that join each other at the places indicated by arrows. SEM backscattered electron image. Adelaide garnet.

A thin section cut normal to the (501) face was also observed with an optical microscope. The wavy lamellae extend from the surface into the interior of the {501} sector. Thicknesses of the wavy lamellae change from place to place, and aluminum-poor lamellae are thicker than aluminum-rich lamellae in almost all areas. Lamellae corresponding to the striations and fine steps are not observed everywhere in the thin section.

A thin section cut normal to (101) terraces on a (601) face was observed under an optical microscope with a tilting stage. The thin section shows growth bands parallel to (101), not to (601). Some growth bands consist of fine lamellae inclined slightly to the band. Thickness of the lamellae is constant in a band, though it varies from less than one micron to several tens of microns throughout the specimen. The fine lamellae, with an interval of less than about 6 μ m, produce iridescence which changes color as the thin section is tilted.

Two kinds of chemically distinct straight lamellae were observed in the section using the electron microprobe. One set of lamellae crosses the wavy lamellae (Fig. 7). The other set of lamellae is commonly joined to the wavy



Fig. 9. Growth bands with lamellae inclined to the bands. Fine lamellae shown by arrows exhibit iridescence. The average distance between the two black lamellae is 1.8 μ m in the area shown by upper arrow and 2.0 μ m in the area indicated by the lower arrow. SEM back-scattered electron image. See text for details. Adelaide garnet.

lamellae on at least one end (Fig. 8). Transformation from the wavy lamellae to the straight lamellae occurs abruptly or gradually. The textures correlate with growth patterns on the surface. The smaller the inclination of the straight lamellae to the growth bands, the shorter the interval between two lamellae; areas with the shorter spacing show iridescence.

In the specimen studied, the shortest distance between two black (or white) lamellae was about 4400Å. Since the thicknesses of black and white lamellae are similar (Fig. 7), the individual lamellae are about 2200Å thick, compared to 1000Å observed with a transmission electron microscope (Hirai and Nakazawa, 1982). These lamellae show iridescence. Figure 9 shows thick straight lamellae without iridescence inclined to the growth bands, and the lamellae are linked to the wavy lamellae between the bands. Also, fine lamellae showing iridescence are observed in Figure 9. Wavy lamellae of the upper-left side of Figure 9 are in another sector.

The iridescent garnets coexist with light colored garnets consisting of well-developed (110) and (112) faces. The dodecahedral garnets do not show any straight or wavy lamellae and are not iridescent.

Discussion

Iridescence in garnets originates from chemically distinct growth lamellae and, as noted, the origin of iridescence is different for the garnets from the two localities. Kamihogi garnet shows simple lamellae parallel to the (110) growth surfaces, and therefore it is concluded that the lamellae result from growth compositional zoning.

The origin of the lamellae in Adelaide garnet is more complicated than that for Kamihogi garnet. Hirai et al. (1982) suggested that wavy and straight lamellae found with the optical microscope and electron microscope were produced by exsolution, not crystal growth, because the wavy lamellae cross the growth zoning, and the oblique straight lamellae are linked to the wavy lamellae. Lamellae crossing growth zoning, however, can be produced in some crystals during growth. In some cases, twins, sectors, and dislocations produced during growth extend through growth zoning parallel to the surface. It is possible that the texture in Adelaide garnet was produced either after growth or during growth.

Hirai et al. (1982) described chemically distinct, coarse lamellae which were believed to have been produced during exsolution. However, they did not think that the fine lamellae producing iridescence were of the same kind as the coarse lamellae, but rather were polysynthetic twins. We have shown above that the thick and thin lamellae have the same character.

The internal textures of Adelaide garnet are correlated with the textures observed on the growth surfaces. The growth surfaces show no evidence of having been etched. Although there is no rigorous proof, the present writers believe that the internal textures corresponding to surface features were probably produced during growth. Adelaide garnet consists mainly of rough faces with striations and small (110) faces. Ingerson and Barksdale (1943) stated that most of the Adelaide garnet occurs in crystals with irregular shapes. The genesis of unusual internal textures and surface features is probably related to the uncommon crystal form, and, therefore, the wavy pattern is rarely observed in garnets with well-developed (110) faces. On the other hand, simple chemical zoning, which in Kamihogi garnet produces iridescence, is observed in grandite garnets from other localities.

Chemical composition may differ between non-equivalent growth sectors of some crystals, such as pyroxene and garnet. For example, the {112} sector is richer in iron than the {110} sector in Kamihogi garnet. This may explain why chemical composition varies at the grooves on the surface. Striations consist of narrow parallel faces. If a face consists of simple growth steps, the sector corresponding to the face will be homogeneous. If the face is striated and consists of alternating microscopic faces with two distinct orientations, the sector may possess lamellae with chemical differences, such as the fine lamellae in Figure 7.

Non-cubic optical properties of grandite garnet have been interpreted in terms of Al/Fe³⁺ ordering (Takéuchi et al., 1982; Akizuki, 1984). Akizuki (1984) suggested that Al/Fe³⁺ ordering is produced metastably during growth and is correlated with the orientations of vicinal faces. Birefringence of the present garnets also can be explained by Al/Fe³⁺ ordering produced on the growth surfaces. The birefringence, fine texture, and iridescence were still visible in specimens from Adelaide which were heated almost to the melting point (Ingerson and Barksdale, 1943). Disordering of Al and Fe³⁺ atoms must occur in the three-dimensional structure during short-term heating. In order to eliminate iridescence, the specimen must be heated for a long time at high temperature such that homogenization occurs.

Garnet from Adelaide consists of many small domains with various degrees of Al/Fe³⁺ ordering. The domain orientations vary with the orientations of the vicinal faces that are inclined to the surface (Akizuki, 1984). This explains the slight difference in orientation of two reciprocal nets observed in X-ray precession photographs taken by Hirai et al. (1982).

In Adelaide garnet, since the interval of some straight lamellae is less than one micron, and the lamellae are nearly parallel to the (110) face, the iridescence is strong in thin sections cut parallel to the lamellae. Since the interval of the lamellae is about 2 μ m in Kamihogi garnet, the iridescence is easily observed in thin sections normal to (110) growth surface. Since the spacing is slightly irregular, iridescence is weak and is not observed on the crystal faces.

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References

- Akizuki, Mizuhiko (1984) Origin of optical variations in grossular-andradite garnet. American Mineralogist, 69, 328–338.
- Dana, E. S. (1920) The System of Mineralogy. Sixth edition. John Wiley & Sons, Inc., New York.
- Hirai, Hisako and Nakazawa, Hiromoto (1982) Origin of iridescence in garnet: An optical interference study. Physics and Chemistry of Minerals, 88, 25–28.

Hirai, Hisako, Sueno, Shigeho and Nakazawa, Hiromoto (1982)

A lamellar texture with chemical contrast in grandite garnet from Nevada. American Mineralogist, 67, 1242–1247.

Ingerson, E. and Barksdale, J. D. (1943) Iridescent garnet from the Adelaide mining district, Nevada. American Mineralogist, 28, 303–312.

Ito, Teiichi (1940) Japanese Minerals in Pictures. Ochi-shoin.

- Sadanaga, Ryoichi and Bunno, Michiaki (1974) The Wakabayashi Mineral Collection. University of Tokyo Press.
- Takéuchi, Yoshio, Haga, Nobuhito, Umizu, Shigetomo, and Sato, Gen (1982) The derivative structure of silicate garnets in grandite. Zeitschrift für Kristallographie, 158, 53–99.

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