Formation of triple-layer coronas between corundum and hornblende from the Lützow-Holm Complex at Akarui Point, East Antarctica

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Triple-layer coronas around corundum occur in the Lützow-Holm Complex at Akarui Point, East Antarctica. The monomineralic layers of green spinel, sapphirine, and plagioclase are arranged in this order from corundum to the matrix hornblende, indicative of the reaction

\[
\text{corundum + hornblende} \rightarrow \text{green spinel + sapphirine + plagioclase + H}_2\text{O-fluid.}
\]

Singular value decomposition analysis in the simplified Na\(_2\)O–CaO–MgO–Al\(_2\)O\(_3\)–SiO\(_2\)–H\(_2\)O system implies that either Na\(_2\)O and CaO, Na\(_2\)O and MgO, or CaO and SiO\(_2\) were open during the corona formation. The position of the initial boundary between corundum and hornblende coincided with the present sapphirine-plagioclase boundary, as judged from the spatial position of the brownish-green spinel in the coronas which is also present in the matrix and hornblende. The flow of Al\(_2\)O\(_3\) decreased outward and completely ceased at the plagioclase-hornblende boundary. This feature together with the monotonic decrease in Al content from corundum to hornblende as well as within sapphirine and plagioclase suggests that diffusion of Al\(_2\)O\(_3\) controlled the corona formation. The net reaction showed an increase in volume, indicating that the products were stable at lower pressures. Therefore, the corona-forming reaction took place during the decompression stage of the Lützow-Holm Complex.

**Keywords**: Corona, Reaction microstructure, Mass transfer, Volume change, Lützow-Holm Complex

INTRODUCTION

Non-equilibrium structures, including coronas, attract the interest of petrologists because of their distinct appearance. Their structural feature that both reactants and products are preserved brings us precious clues for understanding mass transfer and diffusion during metamorphic reactions, as well as pressure (P)–temperature (T) paths (e.g., Nishiyama, 1983; Ashworth and Birdi, 1990; Nakamura, 2002; Santosh and Sajeev, 2006).

An essential approach when studying coronas is the determination of the net reaction of their formation. This requires judging whether the system was open or closed to various components during the reaction. The isocon method proposed by Grant (1986) enables the graphical display of differences in mass of components between the reactants and products. Applying this method to coronas between olivine and plagioclase, Ashworth and Birdi (1990) evaluated the mobility of the components and demonstrated that Al and Si were almost conserved throughout the corona formation. A statistical method, referred to as the singular value decomposition (SVD), provides possible solutions for the net reaction despite substantial analytical errors (Fisher, 1989). Chowdhury et al. (2013) applied the SVD method to double-layered spinel-plagioclase coronas between corundum and hornblende. Four plausible net reactions were proposed based on the textural constraints such as the volume ratio of the spinel and plagioclase, and the occurrence of the spinel as pseudomorphs after corundum by which they assumed no volume change to form the spinel after corundum. All the plausible reactions indicated that Al was almost conserved within the coronas and the neighboring hornblende. The less mobile behavior of Al is often assumed for other coronas around corundum that are ubiquitous in...
low- to high-grade metamorphic belts (e.g., Morishita and Arai, 2001; Santosh and Sajeev, 2006; Raith et al., 2008; Arima et al., 2011).

The determination of the position of the initial boundary between the reactant phases allows us to clarify the growth directions of the layers and the mass transfers across the layer boundaries (e.g., Fukuyama et al., 2006; Mori et al., 2007). Fukuyama et al. (2006) examined the triple-layer reaction zones of diopside, garnet, and wollastonite between metamorphosed mafic dykes and marble. The occurrence of residual calcite within the diopside zone suggested that the initial boundary was set in this zone, and the reasonable growth directions were determined. By performing the two steady-diffusion models for open systems proposed by Ashworth and Birdi (1990) and Johnson and Carlson (1990), they obtained the stoichiometric relations and quantitative mass transfers across the boundaries.

In this study, we dealt with the triple-layer coronas around corundum found in amphibolites from the Lützow-Holm Complex (LHC) at Akarui Point, East Antarctica. We have described the microstructure and chemical compositions and revealed the net reaction of the corona formation and its volume change in an open system. We also determined the position of the initial boundary and the amount of the components that migrated through the layer boundaries as well as the textural evolution. The mineral abbreviations used in this paper are after Whitney and Evans (2010).

**GEOLOGICAL SETTING**

The LHC is a high-grade metamorphic complex along the Prince Olav Coast and Lützow-Holm Bay in East Antarctica. The metamorphic grade increases progressively from the upper amphibolite facies to granulite facies toward the southwest of the complex. The LHC is divided into three metamorphic zones referred to as the amphibolite–facies zone, transitional zone, and granulite–facies zone (Hiroi et al., 1983a, 1987, 1991) (Fig. 1a). The presence of Ca-poor amphiboles and the absence of orthopyroxene characterize the amphibolite–facies zone, while Ca-poor amphiboles are absent, and orthopyroxene is present in the granulite–facies zone. The transitional zone is characterized by the occurrence of both Ca-poor amphiboles and orthopyroxene (Hiroi et al., 1983a; Shiraishi et al., 1984). Kawasaki et al. (2011) estimated the peak $P$-$T$ conditions of Rundvågshetta, the highest-grade area of the LHC, as 1040 °C, 1300–1500 MPa. The subsequent retrograde conditions were 1010 °C, 1000 MPa, and 830 °C, 610 MPa. Symplectites formed by breakdown of garnet, and relic inclusions of kyanite and staurolite in plagioclase and garnet are commonly recognized from some exposures in the LHC, and they are regarded as evidences for a clockwise $P$-$T$ path (e.g., Hiroi et al., 1983a, 1983b, 1986; Kawasaki et al., 1993; Fraser et al., 2000; Iwamura et al., 2013). Sensitive high-resolution ion microprobe (SHRIMP) analyzes yielded metamorphic U–Pb zircon ages of 550–520 Ma (Shiraishi et al. 2003), and electron microprobe U–Th–Pb monazite dating from Skallen showed an age of 560–500 Ma (Hokada and Motoyoshi, 2006).

Akarui Point is an exposure located in the transitional zone (Fig. 1a). It is underlain mainly by garnet-biotite, biotite-hornblende, and hornblende-biotite gneisses with minor metabasites (Yanai et al., 1984) (Fig. 1b). The peak $P$-$T$ conditions for Akarui Point are estimated as follows: 770–980 MPa, 770–790 °C using the garnet-biotite geo-

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**Figure 1.** (a) Geological map of the LHC showing the metamorphic zonation (after Hiroi et al., 1991) and the location of Akarui Point. (b) Geological map of Akarui Point after Yanai et al. (1984) with the sampling locality of the studied amphibolites.
thermometers and garnet-aluminosilicate-silica-plagioclase geobarometers for the silimanite-biotite-garnet gneiss (Kawakami et al., 2008), 825–900 °C using the ternary-feldspar geothermometry for the same gneiss as Kawakami et al. (2008) (Nakamura et al., 2014), 500–600 MPa, 900–920 °C based on the pseudosection combined with the orthopyroxene-spinel geothermometry for the mafic granulite (Iwamura et al., 2013), and 720–840 °C and 720–830 °C using the hornblende-plagioclase geothermometer for the mafic and felsic gneisses (Kazami et al., 2016). Iwamura et al. (2013) proposed the clockwise $P$–$T$ path for Akarui Point as 1100–1200 MPa at ~900 °C for the prograde condition and 500–600 MPa at 900–920 °C for the peak condition. Similar to the metamorphic ages of most areas of the LHC, the SHRIMP zircon U–Pb ages for Akarui Point showed 518 ± 12 Ma (Shiraishi et al., 2003).

SAMPLE DESCRIPTION

The studied amphibolites (sample no. I-022 and TK2002122107B) were collected from different layers of the same outcrop at the Sansyoku Terrace in Akarui Point during the 44th Japan Antarctic Research Expedition (JARE44) (Fig. 1b). The microstructural and petrographic features described below are common to both samples, even though I-022 is orthopyroxene-free while TK2002122107B contains a small amount of orthopyroxene. The amphibolites are composed mainly of hornblende and plagioclase with small amounts of gedrite, corundum, brownish-green spinel, sapphire, biotite and opaque minerals, with or without orthopyroxene. Weak preferred orientation of amphibole grains defines the foliation. Corundum is pink and occurs as flat single crystals approximately 4 mm long. We recognized seven grains of corundum, all of which are surrounded by monomineralic layers of green spinel, sapphire, and plagioclase (Figs. 2a–2j). These layers are arranged in this order from the corundum to the matrix.

Corona

The green spinel layer shows significant variation in thickness that ranges 50–930 µm. The layer is associated with fractures that usually continue to the sapphire layer but do not extend into the plagioclase layer or the corundum (Fig. 2d).

The sapphire layer shows an almost constant thickness of 70–130 µm in contrast to the green spinel and plagioclase layers. It is composed of polycrystalline sapphire with lengths of 0.3–1.7 mm that has an elongation parallel to the layer. Most of the sapphire grains are in contact both with the green spinel and plagioclase layers (Figs. 2a–2c). The sapphire exhibits strong pleochroism from sky-blue to colorless.

The thickness of the plagioclase layer varies significantly (50–330 µm). The layer is composed of polycrystalline plagioclase (250–400 µm long) with the grain boundary almost perpendicular to the layering (Fig. 2c). Most of the plagioclase grains are in contact both with the sapphire layer and the matrix amphiboles (Fig. 2c). The position of optical extinction changes almost perpendicular to the layering, which is due to a gradual change in the chemical composition, as will be described later. Radial cracks occur only around the sapphire layer with a large curvature (Fig. 2e). Some grains exhibit albite twinning. Sporadic grains of brownish-green spinel (60–180 µm in diameter) occur in the plagioclase layer and at the boundary between the sapphire and plagioclase layers (Fig. 2f).

Matrix

The hornblende grains have a maximum length of approximately 1 cm and are bluish-green to brownish-green (Figs. 2g–2i). In some cases, fine-grained (up to 0.5 mm in diameter) and rounded hornblende crystals are included in the matrix plagioclase.

Gedrite (maximum 2 cm long) exhibits pleochroism from brown to pale brown (Figs. 2g and 2i), and the long axis is usually parallel to that of the matrix hornblende grains. There is less gedrite than hornblende in the samples.

Orthopyroxene (maximum 1 mm) is rounded to elongated crystal (Fig. 2i). It is usually not associated with gedrite and vice versa. It exhibits pleochroism from reddish-brown to pale brown.

The plagioclase grains are equigranular (0.7–1.2 mm in diameter) (Figs. 2g–2i). They usually show concentric optical extinction, and some grains also exhibit albite twinning. Polygonal to rounded plagioclase also occurs as inclusions in the matrix hornblende and gedrite grains.

The spinel grains are brownish-green and occur in the matrix and as inclusions in the matrix hornblende and plagioclase (Figs. 2g–2j). They can be easily distinguished by color from the spinel composing the green spinel layer. The spinel in the matrix represents two modes of occurrence. One is rounded and coarse-grained (100–340 µm in diameter) (Figs. 2g and 2h), and the other is elongated and fine-grained (50–100 µm long) (Figs. 2h–2j). The latter is often associated with sapphire that has similar grain sizes and shapes (Fig. 2j). The grain size of spinel occurring as inclusions in the matrix hornblende and plagioclase varies to a large extent (30–350 µm in
Figure 2. Photomicrographs showing the mode of occurrence of the corona (a)-(f) and the matrix (g)-(j) minerals under plane-polarized light except for (c) that is in crossed-polarized light. All images are from I-022 except for (i) that is from TK2002122107B. (a) and (b) Zonal arrangement of the green spinel, sapphirine, and plagioclase. (c) Crossed-polarized light photomicrograph of the same area as (a) showing the polycrystalline plagioclase and sapphirine. (d) Enlargement of (a) showing continuous fractures in both the green spinel and sapphirine. (e) Enlargement of (b) showing the radial cracks in plagioclase developing only around the sapphirine layer with a large curvature. (f) Enlargement of (a) showing the occurrence of brownish-green spinel in the corona. (g) Brownish-green spinel occurring at the grain boundaries of hornblende and as inclusions in hornblende. (h) Equigranular grains of plagioclase with brownish-green spinel and sapphirine occurring at the grain boundaries of the plagioclase. (i) Orthopyroxene as independent grains of gedrite. Brownish-green spinel occurs in the matrix and as inclusions in hornblende. (j) Enlargement of (h) showing the bgSpl + Spr portion. Abbreviations: Bt, biotite; bgSpl, brownish-green spinel; Crn, corundum; Ged, gedrite; gSpl, green spinel; Hbl, hornblende; Pl, plagioclase; Spr, sapphirine; Opq, opaque minerals; Opx, orthopyroxene.
diameter) (Figs. 2g and 2i). It is noted that the brownish-green spinel also occurs in the coronas as inclusions in the plagioclase layer or are in contact with the outer border of the sapphirine layer, as described previously (Fig. 2f).

The sapphirine crystals in the matrix are elongated and fine-grained (50–130 µm long) (Figs. 2h–2j). They are usually associated with the brownish-green spinel described above (Fig. 2j) or occur as inclusions in the matrix plagioclase (Fig. 2h). They exhibit strong pleochroism from sky–blue to colorless.

Biotite (up to 2 mm long) is scattered in the matrix and shows pleochroism from pale yellow to brownish-green (Figs. 2h and 2i). A minor amount of apatite (100 µm in diameter) occurs as inclusions in the gedrite. Opaque minerals usually constitute polygonal aggregates (up to 300 µm) and occur not only in the matrix but also in the corundum, matrix hornblende, and the plagioclase layer (Figs. 2b, 2h, and 2i). The aggregates are composed of pyrite, chalcopyrite, pentlandite, chlorite, and silica minerals. They are partially altered to hydroxides along the fractures and rims except for those in the corundum. The aggregates in the matrix hornblende are connected with the matrix by fractures of the host hornblende which is indicative of ‘pseudo-inclusion’ (cf. Kawakami et al., 2006).

MINERAL CHEMISTRY

The chemical compositions of the constituent minerals in the 1-022 sample were determined using a JEOL JXA-8530F electron probe microanalyzer (EPMA) equipped with wavelength dispersive X-ray spectrometers (WDS) at Kyushu University. Natural and synthetic materials were used for standards. The quantitative analyses were carried out using an accelerating voltage of 15 kV and a beam current of 2 nA for the silicates and oxides, and 10 nA for the sulfides with a probe diameter of 3 µm. The ZAF method was used for data correction. The counting times for the peak and backgrounds were 10 and 5 s, respectively.

The ferric content in spinel and sapphirine was estimated on the basis of the stoichiometry and cation number (cf. Droop, 1987). All iron present in corundum was treated as ferric. Hornblende and gedrite in this study contain less than 10 wt% FeO when all iron was assumed to be ferrous. Application of the method proposed by Holland and Blundy (1994) and Dale et al. (2000) to estimate the ferric content in the amphiboles provided large range, and ferric-free solution was within the range. Therefore, we assume hornblende and gedrite to be free from ferric iron in this study. The representative analyses are shown in Table 1.

Corundum

Corundum is almost pure Al$_2$O$_3$ with small amounts of Fe$^{3+}$ (up to 0.01 apfu based on 3 oxygen, 0.78 wt% of Fe$_2$O$_3$) and Cr (up to 0.01 apfu, 0.38 wt% of Cr$_2$O$_3$).

Sapphirine

Sapphirine shows a variation in $X_{\text{Mg}}$ values of 0.88–0.98 (Fig. 4) and the ferric content less than 1.00 apfu ($O = 40$) irrespective of the mode of occurrence. The coronal sapphirine contains higher Al than the sapphirine occurring in the matrix and as inclusions (Fig. 4). The Cr content of the sapphirine occurring in the matrix and as inclusions are 0.24–0.44 apfu (0.32 apfu in average) and 0.05–0.54 apfu (0.26 apfu in average), respectively. The values are significantly higher than that of the coronal sapphirine (0.00–0.21 apfu, and 0.03 apfu, on average) (Table 1).

In the corona, Al shows a monotonic decrease with increasing distance from the green spinel layer. The Al content in contact with the green spinel (innermost part) ranges 17.77–18.30 apfu, while the Al content next to the plagioclase (outermost part) represents 17.26–18.00 apfu (Fig. 4). Tschermak’s substitution can explain the chemical variations observed in the coronal sapphirine.

Plagioclase

We did not detect K in any of the analyzed plagioclase, and therefore the plagioclase can be regarded as albite–anorthite solid solution. In the corona, the anorthite content [100·Ca/(Ca + Na) in atomic ratio] shows a monotonic decrease with increasing distance from the sapphirine layer. The anorthite content in contact with sapphirine (innermost part) ranges 83–89, while the anorthite content next to the matrix (outermost part) represents 78–84 (Fig. 5). The plagioclase in the matrix represents the anorthite...
content ranging from 81 to 87, and the cores usually have slightly higher anorthite content than the rims (Table 1).

**Hornblende and gedrite**

The amphiboles described as hornblende and gedrite belong to pargasite, and sodic gedrite according to the classification of Leake et al. (1997). They have a similar range of $X_{Mg}$ (0.80–0.83) and Na (0.60–0.80 apfu, O = 23). Most Na in both amphiboles is assigned to the A-site, the largest cation site in the amphibole structure. Hornblende and gedrite have Si of 6.01–6.43 apfu and 5.79–6.15 apfu, respectively, and Ca/(Ca + Na) atomic ratios of 0.67–0.74 and 0.11–0.19, respectively (Fig. 6). The Cr content of the hornblende and gedrite ranges 0.00–0.17 apfu (0.05 apfu in average) and 0.00–0.06 apfu (0.03 apfu in average), respectively.

**Other minerals**

Biotite shows $X_{Mg}$ of approximately 0.90, and TiO$_2$ content of 0.65–0.73 wt%. Pyrite contains small amounts of Co (<1.73 wt%) and Ni (<1.87 wt%). Pentlandite contains small amounts of Cu (<1.64 wt%). Apatite as inclusions in the gedrite belongs to fluorapatite. Chlorite in association with the opaque minerals contains Ni content of 1.11 wt%.
**DISCUSSION**

**Corona-forming net reaction**

The matrix minerals are hornblende, plagioclase, gedrite, brownish-green spinel, sapphirine, biotite and opaque minerals, with or without orthopyroxene, as described previously. We consider that these minerals have equilibrated with corundum before the coronas formed. The brownish-green spinel in the corona has a homogeneous composition which is similar to that in the matrix and as inclusions in the matrix hornblende and plagioclase. This suggests that the brownish-green spinel in the corona was already present before the corona formation.

The microstructural features described previously suggest that the three layers of green spinel, sapphirine, and plagioclase were formed at the expense of corundum and hornblende, expressed as

\[
\text{corundum} + \text{hornblende} = \text{green spinel} + \text{sapphirine} + \text{plagioclase} + \text{H}_2\text{O}-\text{fluid} \tag{R1}
\]

We excluded other minerals present in the matrix for the following reasons: the coronas mostly develop between corundum and hornblende without gedrite or orthopyroxene (Fig. 2b); biotite is also seldom in contact with the coronas, and all the coronal minerals do not contain K; brownish-green spinel and sapphirine in the matrix contain significant amounts of Cr, whereas three minerals composing the layers of the coronas are virtually free from Cr.

The compositions of the reactants and products can...
be treated in the simplified Na₂O-CaO-MgO-Al₂O₃-SiO₂-H₂O system. The chemical compositions of the corundum and green spinel are almost pure Al₂O₃ and spinel (sensu stricto)–hercynite solid solution, respectively. The coronal sapphirine and plagioclase have compositional variations, and their averages were used to estimate the corona-forming reaction. This treatment ignores the movement of any components responsible for the chemical gradients in the sapphirine and plagioclase. The composition of hornblende close to the average of all the analyses was recalculated excluding the minor components (<0.50 wt%). We combined Fe²⁺ with Mg because they can exchange with each other at high temperatures even after corona formation (cf. Nishiyama et al., 2017). Fe³⁺ was treated as Al because the minerals responsible for the corona-forming reaction have trivial amounts of Fe³⁺ that are usually assigned to the same crystallographic site as Al. Therefore, we examine the corona formation in the six-component system of Na₂O-CaO-MgO-Al₂O₃-SiO₂-H₂O, and the following chemical compositions are assigned to each phase: corundum (Crn), Al₂O₃; green spinel (gSpl), MgAl₂O₄; sapphirine (Spr), Mg₆.₈₄Al₁₈.₃₂Si₂.₈₄O₄₀₆; plagioclase (Pl), Ca₀.₈₃Naₐ.₁₇Al₁₈.₃₂Si₂.₈₄O₄₀₆; hornblende (Hbl), Na₀.₆₈Ca₁.₆₅Mg₄.₅₁Al₂.₇₀Si₆.₂₂O₂₅(OH)₂; and H₂O–fluid (W), H₂O.

We applied the SVD method to obtain the stoichiometric relations using the numerical computation software Scilab (developed by Scilab Enterprises). This method gives the stoichiometric relations among minerals by minimizing the residuals of the closed components, where the closed components denote the mass–balanced components among the minerals. In contrast, the components that have inflows or outflows from/to outside of the system during the reaction are referred to as open components. In this study, we evaluated the validity of the assumption of closed or open components by comparing them to the analytical errors, as discussed later.

Only hornblende and plagioclase contain Ca and Na, and they show different ranges for Ca/(Ca + Na) of 0.67–0.74 and 0.78–0.89, respectively. This result requires that at least one of Na₂O and CaO was open during the reaction to obtain the net reaction R₁.

First, we solved the matrices that assumed only either Na₂O or CaO was open, denoted as Case 1 and 2, respectively (Table 2), and obtained the reactions R₂ and R₃ shown in Table 3. Both reactions are inconsistent with the microstructure because either green spinel or sapphirine is represented as reactants.

Second, we examined an additional open component. Previous studies revealed that Al is less mobile compared to other components (e.g., Grant, 1988; Johnson and Carlson, 1990; Ashworth et al., 1992; Ashworth and Sheplev, 1997; Abart et al., 2001; Chowdhury et al., 2013). In this study, the coronal sapphirine and plagioclase show the common feature of outward decrease in Al, as described previously, implying that the Al₂O₃ was less mobile compared to other components. We assumed therefore that Al₂O₃ was the closed component. The combinations of the open components among the remainders are Na₂O and CaO (Case 3), Na₂O and MgO (Case 4), Na₂O and SiO₂ (Case 5), CaO and MgO (Case 6), and CaO and SiO₂ (Case 7), as summarized in Table 2. H₂O–fluid was excluded from the matrices because the stoichiometric coefficient has an identical value and opposite sign to hornblende. The matrices to be solved consist of five phases and three closed components, i.e., rank of three, so we obtain two independent solutions for each case. The linear combination of the two solutions provides a general solution (cf. Nishiyama et al., 2017), which is described as R₆, 9, 12, 15, and 18 in Table 3.

### Table 1. (Continued)

| Mineral | PI | Hbl | Ged | Bt |
|---------|----|-----|-----|----|
| Texture | Matrix | Matrix | Matrix | Matrix |
| Part | RIm | - | - | Bt |
| Point | Pl135 | Amp58 | Amp28 | Bt |
| SiO₂ | 46.23 | 42.55 | 42.43 | 38.39 |
| TiO₂ | 0.00 | 0.20 | 0.18 | 0.65 |
| Al₂O₃ | 32.60 | 15.69 | 18.75 | 17.77 |
| Cr₂O₃ | 0.14 | 0.05 | 0.29 | 0.01 |
| Fe₂O₃ | - | - | - | - |
| Fe²⁺ | 0.10 | 7.20 | 9.18 | 4.67 |
| MnO | 0.07 | 0.19 | 0.21 | 0.10 |
| MgO | 0.01 | 16.65 | 22.46 | 22.79 |
| CaO | 16.71 | 10.54 | 0.85 | 0.00 |
| ZnO | 0.06 | 0.00 | 0.05 | 0.27 |
| Na₂O | 2.16 | 2.42 | 2.59 | 1.01 |
| K₂O | 0.08 | 0.34 | 0.02 | 8.87 |
| Total | 98.16 | 95.83 | 97.01 | 94.53 |

O= 8 23 23 22
Si 2.17 6.19 5.97 5.48
Ti 0.00 0.02 0.02 0.07
Al 1.80 2.69 3.11 2.99
Cr 0.01 0.01 0.03 0.00
Fe²⁺ - - - -
Fe³⁺ 0.00 0.88 1.08 0.56
Mn 0.00 0.02 0.02 0.01
Mg 0.00 3.61 4.71 4.85
Ca 0.84 1.64 0.13 0.00
Zn 0.00 0.00 0.00 0.03
Na 0.20 0.68 0.71 0.28
K 0.00 0.06 0.00 1.62
Total 5.02 15.80 15.78 15.89
We obtained the ranges of the variable so that the solutions have proper signs for the stoichiometric coefficients consistent with the microstructure, that is, $0.30 < p < 0.64$ (R6), $0.39 < q$ (R9), $0.31 < r < 0.67$ (R12), and $0.27 < t < 0.53$ (R18). The reaction R15 is always inconsistent with the microstructure at any value of $s$.

As mentioned previously, we evaluated the validities of the closed and open components by comparing their residuals to the propagated analytical errors. That is, when the assumption of the closed and open components is valid, the residuals for each component should be smaller than the analytical errors for the closed components and larger than the errors for the open components. These relations are expressed as

For closed components

$$
\sum_{i} n_i < \sum_{i} (v_i N_i \sigma_i)^2
$$

(Eq. 1)

For open components

$$
\sum_{i} n_i > \sum_{i} (v_i N_i \sigma_i)^2
$$

(Eq. 2)

for the calculations are listed in Table 4. As a result, all the components in R9, 12, and 18 satisfy both criteria within the reasonable ranges for $q$, $r$, and $t$, respectively. However, CaO does not satisfy the criterion for the open component in the range of $0.63 < p < 0.64$ for R6. We excluded this range and concluded that $0.30 < p < 0.63$ is the plausible range for $p$.

### Mass transfer and textural evolution

The brownish-green spinel is absent in the corundum, and also in the green spinel and sapphirine layers, whereas it does occur in places outside of the sapphirine layer, as described previously. This suggests that the space of the coronas where the brownish-green spinel exists was occupied by the matrix hornblende before the corona formation. We therefore assume that the position of the initial boundary between corundum and hornblende corresponds to the present boundary between the sapphirine and plagioclase layers. Another possibility is that the initial boundary was located further inside of the coronas and will be discussed later.

The present assumption suggests that sapphirine and plagioclase were not produced or consumed at the sapphirine–plagioclase boundary. This requires that sapphirine grew inward and plagioclase grew outward (black arrows in Fig. 7). The presence of the green spinel layer on inner side of the sapphirine layer indicates the growth of sapphirine at the expense of green spinel that simultaneously started to form at the initial boundary. In other words, the green spinel layer grew inward at the expense of corundum and was replaced by sapphirine at its outer boundary at the same time.

These interpretations enable the division of the net reactions R6, 9, 12, and 18 into three local reactions (I–IV).
(III) that took place at the layer boundaries with the exception of the sapphire–plagioclase boundary.

(I) Plagioclase was produced at the expense of hornblende at the boundary between hornblende and plagioclase. This reaction in Cases 3, 4, 5, and 7 is described as:

\[
\text{Hbl} + (-2.13p + 1.37)\text{CaO} + (-2.35p + 1.98)\text{Al}_2\text{O}_3 \\
+ (-2.57p + 3.64)\text{Pl} + W + (5.57p - 1.68)\text{SiO}_2 \\
+ 4.51\text{MgO} + (0.22p + 0.03)\text{Na}_2\text{O} \quad (\text{R6-1}),
\]

and

\[
\text{Hbl} + 2.31\text{Al}_2\text{O}_3 + 1.67\text{CaO} + (7.15t + 0.53)\text{SiO}_2 \\
= 4.00\text{Pl} + W + 4.51\text{MgO} + (7.15t - 1.93)\text{SiO}_2 \quad (\text{R18-1}),
\]

respectively. Note that the components expressed on the left-hand side are supplied by other local reactions or from outside of the system, and those on the right-hand side are released toward other boundaries or outside of the system.

(II) The reactions to produce green spinel at the boundary between corundum and green spinel are expressed for Case 3, 4, 5, and 7 as:

\[
\begin{align*}
\text{Hbl} + (11.71q - 4.54)\text{MgO} + 0.47\text{Al}_2\text{O}_3 \\
= 1.99\text{Pl} + W + (11.71q - 0.03)\text{MgO} \\
+ 0.17\text{Na}_2\text{O} + 1.91\text{SiO}_2 \quad (\text{R9-1}),
\end{align*}
\]

\[
\begin{align*}
\text{Hbl} + 0.47\text{Al}_2\text{O}_3 &= 1.99\text{Pl} + W + 4.51\text{MgO} \\
+ (5.11t - 1.57)\text{SiO}_2\text{outward} + 0.17\text{Na}_2\text{O} \\
+ (-5.11t + 3.47)\text{SiO}_2\text{outward} \quad (\text{R12-1}),
\end{align*}
\]

and

\[
\begin{align*}
\text{Hbl} &+ 1.17\text{H}_{2}\text{O-fluid} \\
&= 1.76\text{CaO} + (7.15t + 0.53)\text{SiO}_2 \\
&+ 0.31\text{H}_{2}\text{O}\text{outward} + 0.17\text{Na}_2\text{O} \\
+ (-5.11t + 3.47)\text{SiO}_2\text{outward} \quad (\text{R12-1}),
\end{align*}
\]

respectively. Note that the components expressed on the left-hand side are supplied by other local reactions or from outside of the system, and those on the right-hand side are released toward other boundaries or outside of the system.

Note that the components expressed on the left-hand side are supplied by other local reactions or from outside of the system, and those on the right-hand side are released toward other boundaries or outside of the system.

### Table 3. Stoichiometric coefficients of the phases and residual amounts of the open components

| Case | Open component(s) | Crm | Hbl | gSpr | Spr | Pl | W | Open component(s) |
|------|-------------------|-----|-----|------|-----|----|---|------------------|
| 1    | Na$_2$O           | -6.54 | -1  | -0.08 | 0.67 | 1.99 | 1 | 0.17Na$_2$O (R2) |
| 2    | CaO               | -4.81 | 10.43 | -0.87 | 4.00 | 1 | -1.67CaO (R3) |
| 3    | Na$_2$O and CaO   | -7.32 | -1  | -4.86 | 1.37 | 1.07 | 1 | 0.25Na$_2$O+0.76CaO (R4) |
|      |                   | -5.12 | 8.55 | -0.59 | 3.64 | 1 | 0.03Na$_2$O+1.37CaO (R5) |
|      |                   | -2.20p+5.12 | -1 | -13.41p+8.55 | 1.96p-0.59 | -2.57p+3.64 | 1 | (0.22p+0.03)Na$_2$O (R6) |
|      |                   |       |     |       |     |     |   | (2.13p-1.37)CaO |
| 4    | Na$_2$O and MgO   | -13.71 | -1  | 7.09 | 0.67 | 1.99 | 1 | 0.17Na$_2$O-7.17MgO (R7) |
|      |                   | -2.00 | -1  | -4.62 | 0.67 | 1.99 | 1 | 0.17Na$_2$O+4.54MgO (R8) |
|      |                   | -11.71q-2.00 | -1 | 11.71q-4.62 | 0.67 | 1.99 | 1 | 0.17Na$_2$O (R9) |
|      |                   |       |     |       |     |     |   | (+11.71q+4.54)MgO |
| 5    | Na$_2$O and SiO$_2$ | -7.88 | -1  | -4.03 | 1.25 | 1.99 | 1 | 0.17Na$_2$O-1.64SiO$_2$ (R10) |
|      |                   | -3.70 | -1  | 8.28 | -0.55 | 1.99 | 1 | 0.17Na$_2$O+3.47SiO$_2$ (R11) |
|      |                   | -4.18r-3.70 | -1 | -12.32r+8.28 | 1.80r-0.55 | 1.99 | 1 | 0.17Na$_2$O (R12) |
|      |                   |       |     |       |     |     |   | (+5.11r+3.47)SiO$_2$ |
| 6    | CaO and MgO       | 8.72  | -1  | -3.09 | -0.87 | 4.00 | 1 | -1.67CaO+13.53MgO (R13) |
|      |                   | -0.03 | -1  | 5.66 | -0.87 | 4.00 | 1 | -1.67CaO+4.78MgO (R14) |
|      |                   | 8.75s-0.03 | -1 | 8.75s+5.66 | -0.87 | 4.00 | 1 | -1.67CaO (R15) |
|      |                   |       |     |       |     |     |   | (+8.75s+4.78)MgO |
| 7    | CaO and SiO$_2$   | -11.08 | -1  | -8.06 | 1.84 | 4.00 | 1 | -1.67CaO-7.68SiO$_2$ (R16) |
|      |                   | -5.24 | -1  | 9.16 | -0.68 | 4.00 | 1 | -1.67CaO-0.53SiO$_2$ (R17) |
|      |                   | -5.84s-5.24 | -1 | -17.22s+9.16 | 2.52s-0.68 | 4.00 | 1 | -1.67CaO (R18) |
|      |                   |       |     |       |     |     |   | (+7.15s-0.53)SiO$_2$ |

All stoichiometric coefficients were normalized to hornblende. The stoichiometric coefficients of the reactants and products are shown with negative and positive signs, respectively. The linear combinations were calculated as (R6) = p(R4) + (1−p)(R5), (R9) = q(R7) + (1−q)(R8), (R12) = r(R10) + (1−r)(R11), (R15) = s(R13) + (1−s)(R14), (R18) = t(R16) + (1−t)(R17), where p, q, r, s, and t were arbitrary numbers. Abbreviation W represents H$_2$O-fluid, others are same as in Table 1.

### Table 4. Standard deviations for the components derived from counting errors of the EPMA analysis (1σ in %)

| Component(s) | Crm | Hbl | gSpr | Spr | Pl |
|--------------|-----|-----|------|-----|----|
| SiO$_2$      | 0.55 | 0.55 | 1.12 | 0.53 |    |
| Al$_2$O$_3$  | 0.34 | 0.94 | 0.46 | 0.45 | 0.59 |
| MgO          | 0.85 | 0.85 | 0.80 |      |    |
| CaO          | 1.36 |      |      | 1.11 |    |
| Na$_2$O      | 2.62 |      |      | 2.75 |    |

Abbreviations are same as in Table 1.
The reactions that took place at the boundary between green spinel and sapphirine for Cases 3, 4, 5, and 7 are:

\[(11.71q + 2.00)\text{Crn} + (11.71q - 0.03)\text{MgO} = (11.71q - 0.03)\text{gSpl} + 2.03\text{Al}_2\text{O}_3\]  
\[(4.18r + 3.70)\text{Crn} + 4.51\text{MgO} = 4.51\text{gSpl} + (4.18r - 0.81)\text{Al}_2\text{O}_3\]

and

\[(5.84t + 5.24)\text{Crn} + 4.51\text{MgO} = 4.51\text{gSpl} + (5.84t + 0.73)\text{Al}_2\text{O}_3\]

respectively.

(III) The reactions that took place at the boundary between green spinel and sapphirine for Cases 3, 4, 5, and 7 are:

\[(2.20p + 5.12)\text{Crn} + 4.51\text{MgO} = 4.51\text{gSpl} + (2.20p + 0.61)\text{Al}_2\text{O}_3\]  
\[(13.41p - 4.04)\text{gSpl} + (2.20p + 0.61)\text{Al}_2\text{O}_3 + (5.57p - 1.68)\text{SiO}_2 = (1.96p - 0.59)\text{Spr} + (-2.35p + 1.98)\text{Al}_2\text{O}_3\]

\[(12.32r - 3.77)\text{gSpl} + (4.18r - 0.81)\text{Al}_2\text{O}_3 + (5.11r - 1.57)\text{SiO}_2 = (1.80r - 0.55)\text{Spr} + 0.47\text{Al}_2\text{O}_3\]

respectively.
The local reaction R12–1 for Case 5 shows that SiO₂ moves toward both directions from the boundary between the plagioclase layer and hornblende. If all the SiO₂ moves inward, green spinel would be completely replaced by sapphirine. The bidirectional flows are artificial in order to leave green spinel. Therefore, we discard Case 5, and conclude that Cases 3, 4, and 7 are plausible combinations of the open components for the corona formation.

The examples in Figure 7 illustrate the amounts of transferred components and minerals that were consumed and produced at the boundaries when \( p, q, \) and \( t \) are 0.45, 0.65, and 0.35, respectively. The features such as direction of mass transfer and position of complete consumption are independent of their values even the amounts of mass transfer vary. The directions of boundary movements are also unchanged. We obtained the following common features among Cases 3, 4, and 7. Green spinel formed at the inner boundary was set at a position inner to the boundary between sapphirine and plagioclase. The radial cracks in the plagioclase layer, the green spinel as spinel, the sapphirine as sapphirine layers (Fig. 2d). Similar crystal structures between spinel and sapphirine of cubic close-packed oxygen (cf. Moore, 1968, 1969) could be responsible for the preservation of the fractures in the sapphirine after the green spinel. The fractures usually penetrate the full width of the green spinel and sapphirine layers (Fig. 2d). Provided that the initial boundary was set at a position inner to the boundary between sapphirine and plagioclase, two reactions are required to produce a single layer of green spinel or sapphirine depending on the position of the initial boundary. This means that a layer of green spinel or sapphirine would grow on both sides. It is unlikely that such a mineral would possess the common fractures that continue in full width of both layers as observed in the samples.

MgO supplied from hornblende and outside of the system, and Al₂O₃ released from corundum reach the entire distance of the corona. The flux of Al₂O₃ decreases with increasing distance from corundum, while that of MgO is constant until the MgO meets corundum. Corundum centered in the coronas is almost pure Al₂O₃, and all corona–constituting minerals and hornblende contain Al as major elements. The Al content of these minerals decreased monotonically outward in the following order: corundum (26.55–26.43, normalized by 40 oxygen), green spinel (19.79–19.21), sapphirine (18.30–17.26), plagioclase (9.80–8.84), and hornblende (4.66–3.81). In addition, the coronal sapphirine and plagioclase show an outward decrease in Al content within the layers, as mentioned previously. These features suggest that the corona formation and zonal arrangement were controlled by the diffusion of Al₂O₃.

### Table 5. Molar volume of minerals after Holland and Powell (1998)

| Phase     | Formula | Molar volume [J/bar] |
|-----------|---------|----------------------|
| Corundum  | Al₂O₃   | 2.558                |
| Spinel    | MgAl₂O₄ | 3.978                |
| Sapphirine| Mg₅Si₃Al₂SiO₁₂ | 19.775            |
| Anorthite | CaAl₂Si₂O₈ | 10.079           |
| Pargosite | NaCa₂Mg₃Al₄Si₄O₁₄(OH)₂ | 27.190         |

### Table 6. Net and local volume changes associated with the corona-forming reaction

| Open components | \( \Delta V_{\text{mineral}} \) | \( \Delta V_{\text{Crn} \rightarrow \text{gSpl} + \text{Spr}} \) | \( \Delta V_{\text{Hbl} \rightarrow \text{Pl}} \) |
|-----------------|-------------------------------|---------------------------------|---------------------|
| Na₂O and CaO    | +12\(\%\)                     | +21\(\%\)                      | +6\(\%\)           |
| Na₂O and MgO    | +6\(\%\)                      | +57\(\%\)                      | -26\(\%\)          |
| CaO and SiO₂    | +31\(\%\)                     | +3\(\%\)                       | +48\(\%\)          |

\( \Delta V_{\text{mineral}} = 100 \cdot \left[ (V_{\text{gSpl}} + V_{\text{Spr}} + V_{\text{Pl}}) - (V_{\text{Hbl}} + V_{\text{Crn}}) \right]/(V_{\text{Hbl}} + V_{\text{Crn}}). \)

\( \Delta V_{\text{Crn} \rightarrow \text{gSpl} + \text{Spr}} = 100 \cdot (V_{\text{gSpl}} + V_{\text{Spr}} - V_{\text{Crn}})/V_{\text{Crn}}. \)

\( \Delta V_{\text{Hbl} \rightarrow \text{Pl}} = 100 \cdot \left( V_{\text{Pl}} - V_{\text{Hbl}} \right)/V_{\text{Hbl}}. \)

Abbreviations are same as in Table 1.

### Volume change and timing of the corona-forming reaction

We estimated the net volume changes between the reactant and product minerals, \( \Delta V_{\text{mineral}} \), derived from net reactions R6, 9, and 18 based on the dataset of Holland and Powell (1998), considering the hornblende as pargasite, the green spinel as spinel, the sapphirine as sapphirine (793), and the plagioclase as anorthite, as shown in Table 5. As a result, the calculated \( \Delta V_{\text{mineral}} \) for all three cases show positive values (Table 6).

We also calculated the local volume changes from hornblende to plagioclase (\( \Delta V_{\text{Hbl} \rightarrow \text{Pl}} \)) and from corundum to green spinel and sapphirine (\( \Delta V_{\text{Crn} \rightarrow \text{gSpl} + \text{Spr}} \)). The latter are positive in all cases (Table 6). These estimations indicate that the positive \( \Delta V_{\text{mineral}} \) is mainly ascribed to the growth of the green spinel and sapphirine layers. Therefore, the inner two layers expanded against the plagioclase layer. The radial cracks in the plagioclase layer around the sapphirine layer with a large curvature (Fig. 2e), where stress can concentrate, are interpreted to be the result of volume increase.
CONCLUSIONS

The following features were revealed in this study. The coronas were formed by open-system net reactions expressed as corundum + hornblende = green spinel + sapphire + plagioclase + H₂O-fluid with open components either of Na₂O and CaO, Na₂O and MgO, or CaO and SiO₂. These reactions are associated with an increase of the net volume, suggesting that the reactions took place during decompression. The position of the initial boundary between corundum and hornblende corresponds to the present boundary between the sapphireine and plagioclase layers. This constraint enables the determination of the amount of mass transfer across the boundaries and suggests that the diffusion of Al₂O₃ controlled the rate of mass transfer.

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