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A cooling rate constraint on microtextural development of plagioclase and scapolite: an example from the Lützow-Holm Complex, East Antarctica

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Abstract: Exsolution lamellae in albite-rich plagioclase and antiphase domains in scapolite from the Lützow-Holm Complex of East Antarctica were found under a transmission electron microscope. These micro-textures were found in the first cooling period of the three period cooling model proposed from the geochronological data. Based on these micro-textures, the cooling rate of the complex was estimated to be in a range from several to thousands K/my. These rates are concordant with those estimated from the ages of the complex.

key words: cooling rate, exhumation rate, Lützow-Holm Complex, microtexture, transmission electron microscope analysis

1. Introduction

The Lützow-Holm Complex, which extends along the Prince Olav Coast and Lützow-Holm Bay region, is characterized by westward progressive metamorphism from the upper amphibolite facies to granulite facies and a well-documented clockwise *P-T-t* path of these rocks (*e.g.* Hiroi *et al.,* 1991). K/Ar, ^{40}Ar/^{39}Ar and U/Pb zircon ages have been reported from the Lützow-Holm complex by several previous studies (*e.g.* Kaneoka *et al.,* 1968; Shiraishi *et al.,* 1994; Fraser and McDougall, 1995; Fraser *et al.,* 2000).

There are a few approaches to estimate the cooling rates of the metamorphic rocks, which give a basis for studying the exhumation of the rocks. One approach is that of thermochronology, which utilizes isotopic ages derived from minerals with different closure temperatures. By comparing K/Ar and ^{40}Ar/^{39}Ar ages, Fraser and McDougall (1995) estimated the cooling rate of the Lützow-Holm Complex is several to tens K/My. Recently, Fraser *et al.* (2000) suggests a cooling and exhumation history with three periods and estimated the exhumation rates in the first cooling period.

Another approach for estimating the cooling rate is a study of microstructures in constituent minerals. For example, the size of antiphase domain (APD) and the width of exsolution lamellae in pigeonite and plagioclase feldspar have been used to study the thermal history of various rocks such as meteorites (*e.g.*, Kitamura *et al.,* 1983; Weinbruch and Müller, 1995) and igneous rocks (*e.g.*, Miyake and Shimobayashi, 2000).

The micro-textures of the constituent minerals of the Lützow -Holm Complex were studied under a transmission electron microscope (TEM). We report exsolution texture in plagioclase and antiphase domain structure in scapolite and estimate the cooling rate of the complex based on these micro-textures and a three periods cooling model by Fraser *et al.* (2000).

2. Micro-textures in plagioclase and scapolite

Specimens of plagioclase (~An_{15}) were selected from a rock chip (*Sp. 81012011*) from Akebono Rock. Specimen of scapolite was selected from a rock chip (*Sp. 81020302C*) from Langhovde. The composition of the scapolite specimens is $Na_{1.14}Ca_{2.83}Al_{4.80}0Si_{7.20}Cl_{0.25}$ $(CO_3)_{0.87}$, which can be expressed as Me_{71}, where the amount of CO_3 is calculated based on the charge balance. Both specimens are chemically homogeneous under back-scattered electron images. Both plagioclase and scapolite specimens were removed from petrographic thin sections and sandwiched by Cu grids of 3 mm diameter for TEM study. They were further thinned by ion beam bombardment. Observations were made with a HITACHI H-8000 microscope at Kyoto University operating at 200 kV.

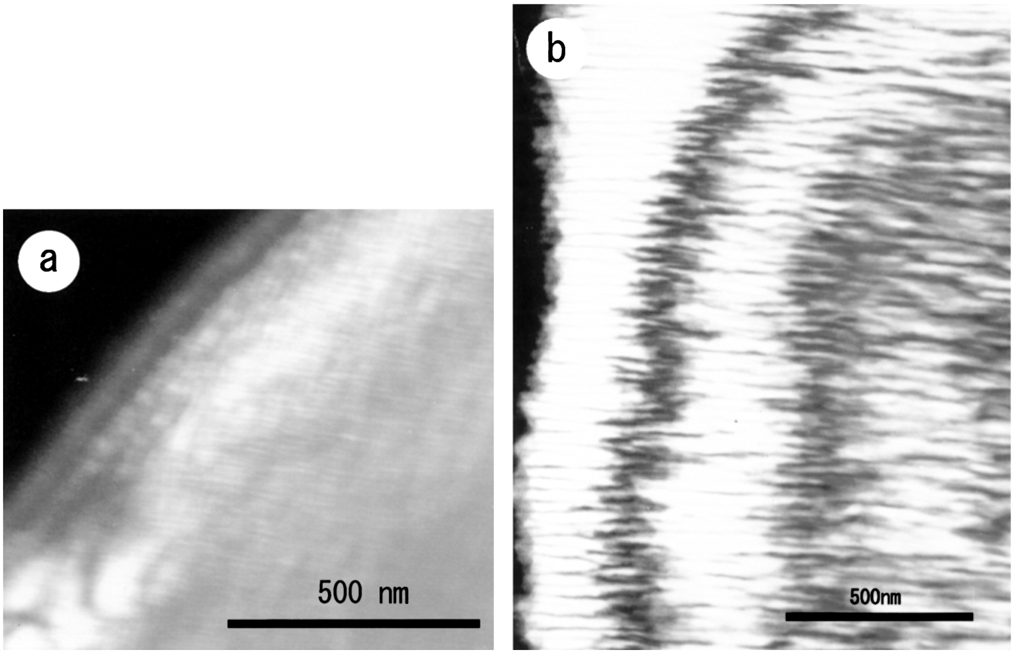

Fig. 1. Transmission electron micrograph of the lamellar intergrowth in albite-rich plagioclase from Akebono Rock (Sp. 81012011). The compositional fluctuations produce fluctuations in local lattice spacings and result in periodic fluctuations in the diffraction contrast. Wavelength is the thickness of these periodic fluctuations, (a) wavelength: 13 nm. (b) wavelength: 30 nm

2.1. Exsolution lamellae in Na-rich plagioclase

Lamellar intergrowths of albite and oligoclase, caused by the peristerite miscibility gap, were observed in plagioclase under TEM (Fig. 1). The lamellae look like having been developed from periodic compositional fluctuations and often slightly waved, indicating the formation by the spinodal decomposition. The average wavelengths of the lamellar texture of two grains were measured as approximately 13 and 30 nm, respectively. One possible reason as to why two grains from same rock chip had different wavelength, is thought that each area has a little compositional difference, but the difference cannot be detected using our system. Since the wavelength in the lamellar texture varies to some extent, we take the range from 13 and 30 nm as the wavelength in further study.

2.2. Antiphase domains in scapolite

Scapolite has two polymorphs: a high temperature form with *I*-lattice and a low temperature form with *P*-lattice (Phakey and Ghose, 1972). Previous researchers (Phakey and Ghose, 1972; Hassan and Buseck, 1988) reported the presence of antiphase domain boundaries (APB's) in intermediate composition of scapolite. *P*-lattice symmetry gives two types of reflections: type *a* ($h+k+l=$even) and type *b* ($h+k+l=$odd) reflections, while *I*-lattice gives only type *a* reflection. The presence of APB's indicates

*Fig. 2. Dark-field (**g**=423) electron micrograph of antiphase domains in scapolite from Langhovde (Sp. 81020302C). The average size of fine domains was 50×50 nm^2.*

the transition from *I*-lattice symmetry to *P*-lattice symmetry.

The present specimens show *P*-lattice symmetry under TEM. APB's were observed by using type *b* reflections as shown in Fig. 2, indicating the transition from high to low temperature forms. The average size of domains was measured as about 50×50 nm^2 or less.

3. Estimation of cooling rate from lamellar texture in plagioclase

The wavelengths of lamellar texture have been known to be strongly related to annealing duration or cooling rates. For example, the relation between the wavelength and the annealing duration in the exsolution of clinopyroxene is determined by experiments on the spinodal decomposition process, *i.e.,* phase separation process (*e.g.*, McCallister, 1978). Based on such relations, the relation between the wavelength and the cooling rate can also be obtained. On the other hand, annealing experiments for the homogenization processes of the lamellar texture caused by the peristerite miscibility gap have been carried out to estimate the coefficients for interdiffusion of NaSi-CaAl in peristerite (Yund, 1986; Liu and Yund, 1992). In order to estimate the cooling rate from the lamellar texture in the present paper, therefore, we simply assume that the change of the wavelength of the exsolution lamellae is the same both in phase separation and homogenization processes and is controlled by the diffusion of elements in the crystal.

The annealing experiments of plagioclase (Yund, 1986; Liu and Yund, 1992) show that half of the wavelength of the lamellae (l) can be expressed as;

$$l^2 = 2D \cdot t, \tag{1}$$

where t is the annealing duration at a certain temperature and D the average interdiffusion coefficients. Equation (1) has the same form as the mean diffusion distance in the one dimensional case. The diffusion coefficient is expressed as;

$$D(t) = D_0 e^{-Q/RT}, \tag{2}$$

where D_0 is the pre-exponential factor, Q the activation energy and R the gas constant. The average interdiffusion coefficients for NaSi-CaAl exchange in plagioclase for the peristerite interval (~An_0 to ~An_{26}) were estimated by the lamellar homogenization experiments under the conditions where pressure is 1.5 GPa, temperature 1323 to 1173 K, ~1.0 wt% H_2O added, and hydrogen fugacity fixed at the magnetite/hematite buffer (Liu and Yund, 1992), and where pressure is 1.5 GPa, temperature 1323 to 1173 K, ~1.0 wt% H_2O added, and no buffer used (Yund, 1986). These two experiments give an activation energy (Q) for CaAl-NaSi interdiffusion of 303 kJ/mol (Liu and Yund, 1992) and 405 kJ/mol (Yund, 1986) and a pre-exponential factor (D_0) of $3 \times 10^{-8} m^2/s$ (Liu and Yund, 1992) and $1.8 \times 10^{-4} m^2/s$ (Yund, 1986), respectively. The diffusion coefficient at the temperature for the exsolution phenomena was obtained by extrapolation of the data obtained at 1323–1173 K. This extrapolation creates uncertainty associated with the diffusion coefficient. In the present paper, we employ both diffusion coefficients by Yund (1986) and Liu and Yund (1992) to reduce the inaccuracy caused by the extrapolation.

Fraser *et al.* (2000) proposed the three periods exhumation and cooling history for Rundvägshetta, the first is exhumation period for 20 my during which rocks exhume and cool from about 35 km and >1173 K to about 14 km and 623 K (stippled area in Fig. 8); the second is no exhumation at about 12 km depth and 573 K for about 100 my; the last is further exhumation and cooling period.

Diffusion lengths of cations in plagioclase at the temperature in the second period, 573 K for 100 my, are 3.6×10^{-1} nm and 1.1×10^{-3} nm from eqs. (1) and (2) using the diffusion coefficients (Liu and Yund, 1992; Yund, 1986), respectively. These values indicate that the second period had little influence on the developments of exsolution lamellae in plagioclase. Similarly, the cooling in the third period, which is lower temperature than the second period, can be shown to have no significant effect to the lamellae coarsening. Therefore, the formation event of exsolution lamellae in plagioclase can be assigned to only the first period.

Equation (1) for diffusion at constant temperature can be rewritten to include the temperature change during the diffusion process as;

$$l^2=2\int_0^t D(t)\,\mathrm{d}t. \tag{3}$$

Here, we simply assume an exponential cooling rate in the first period as;

$$T(t)=T_e e^{-t/\tau}, \tag{4}$$

where T_e is the temperature of the spinodal curve at certain composition and τ the cooling time scale, *i.e.,* the duration within which the temperature decreased to $1/e$ of T_e (K). By substituting eq. (2) and eq. (4) into eq. (3) and integrating for $t\rightarrow\infty$, we obtain the relation between the length scale (l) and the cooling time scale (τ) as;

$$\tau=\frac{Ql^2}{2RT_eD_0e^{-Q/RT_e}}. \tag{5}$$

Then, by using eqs. (4) and (5), the cooling rate ($\mathrm{d}T/\mathrm{d}t$) at T_e can be expressed by l as;

$$\left.\frac{\mathrm{d}T}{\mathrm{d}t}\right|_{t=0}=-\frac{2T_\mathrm{e}^2RD_0}{Ql^2}e^{-Q/RT_e}. \tag{6}$$

Here we assume an initial temperature (T_e) of 773 K for the start of the spinodal decomposition process with the composition An_{15} (Carpenter, 1994). The cooling rates were calculated by eq. (6) for two diffusion coefficients and two wavelengths. The cooling rates is in the range from -2.4×10^3 to -2.6 K/my for the range of the wavelength at 773 K. The cooling rates calculated are listed in Table 1.

Table 1. Calculation of cooling rate at T_e (773K) based on equation and assumptions discussed in the text. dT/dt (L&Y) is the values using the experiment data of Liu and Yund (1992) and dT/dt (Y) using data of Yund (1986).

Wave length (nm)	$\mathrm{d}T/\mathrm{d}t$ (L&Y) (K/My)	$\mathrm{d}T/\mathrm{d}t$ (Y) (K/My)
13	$\sim-2.4\times10^3$	$\sim-1.4\times10^1$
30	$\sim-4.6\times10^2$	~-2.6

4. Comparison with other cooling rate

The cooling rate was estimated at about $-30\ [=-(>1173-623\,\mathrm{K})/20\,\mathrm{my}]$ K/my on the average in the first cooling period from Fig. 8 (the stippled area) in Fraser *et al.* (2000). Fraser and McDougall (1995) also calculated the average cooling and exhumation rates from K/Ar and ^{40}Ar/^{39}Ar mineral ages of several localities in the Lützow-Holm Complex. They suggest the rates of several to several tens K/my when the P-T condition changed from 973–1173 K and 0.55–1.0 GPa to 573 K and 0.4 GPa. The values of the cooling rate estimated in the present study, several to thousands K/my at 773 K, are consistent with those values, about ten to 30 K/my estimated at 773 K, suggested by Fraser and McDougall (1995) and Fraser *et al.* (2000), although these values of both rates were estimated with the quite different methods. Especially, the cooling rate estimated using the unbuffered diffusion coefficient (Yund, 1986), several to ten K/my, is very similar to those values. This suggests the possibility that the H_2, O_2, and/or H_2O fugacities in Lützow-Holm Complex were under the similar condition in

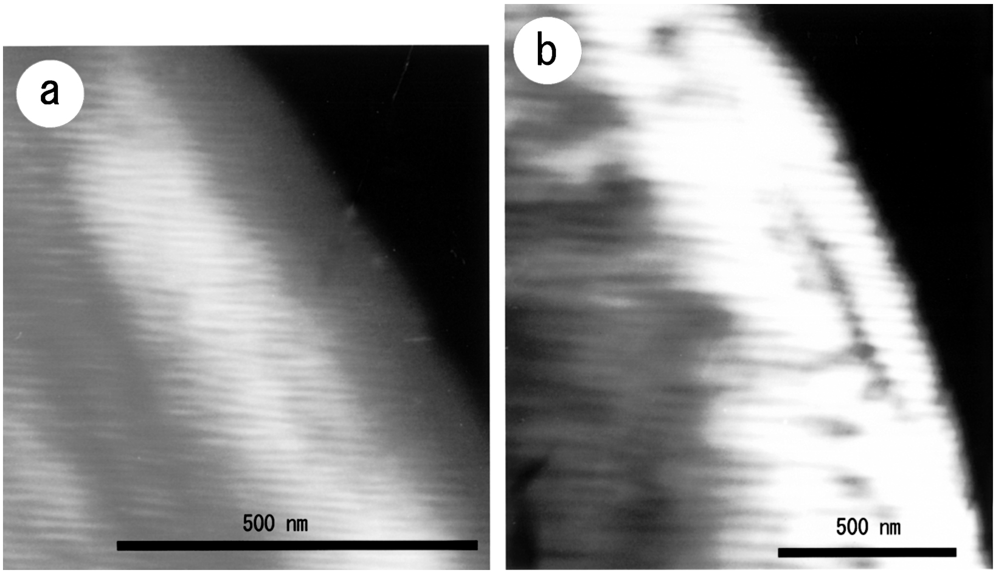

Fig. 3. Transmission electron micrograph of the lamellar intergrowth in albite-rich plagioclase from Sanbagawa belt. (a) Wavelength: 17 nm. (b) wavelength: 42 nm

Table 2. Calculation of cooling rate of Sanbagawa belt based on assumptions discussed in the text. The values (L&Y) are the values with the experiment data by Liu and Yund (1992) and (Y) by Yund (1986).

Wavelength (nm)	dT/dt (L&Y) (K/My)	dT/dt (Y) (K/My)
17	$\sim -1.4\times10^3$	~ -8.0
42	$\sim -2.4\times10^2$	~ -1.3

the unbuffered experiment.

The relative cooling rate can also be discussed by comparing the exsolution texture with those in other metamorphic rocks. Figure 3 shows micrographs of exsolution lamellae in plagioclase ($\sim An_{15}$) in the Besshi area of the Sanbagawa belt, central Shikoku (sample locality: locality A in Shirahata and Hirajima, 1995) and the wavelength is in the range from 17 to 42 nm, which correspond to a cooling rate in the range from -1.4×10^3 to -2.4×10^2 K/my if the initial temperature is 773 K (Table 2). Then, the cooling rate at 773 K of the Lützow-Holm Complex (Table 1) is similar to or a little faster than that of Sanbagawa belt. The cooling history of the Lützow-Holm Complex can also be compared with other metamorphic rocks by using the APD size observed in scapolite. The APD, *e.g.*, type b-APD in anorthite-rich plagioclase (Carpenter, 1991), has also been known to coarsen in proportion to annealing time and cooling rate. Although we cannot estimate the cooling rate from the size of APD in scapolite because of no experimental data of the activation energy and others, scapolite of the Lützow-Holm Complex (Fig. 2) has smaller APD's than those observed in the specimens from other localities: about 6000 nm^2 in metamorphic carbonate rocks from Central Alps (Oterdoom and Wenk, 1983) and 1 to 3 μm^2 from pegmatite at Gouverneur, New York (Me_{37}) (Phakey and Ghose, 1972). The differences in the size of APD may also suggest the complex cooled faster than other metamorphic rocks and pegmatite.

5. Conclusion

The lamellar texture in plagioclase was formed in the first period proposed by Fraser *et al.* (2000). The cooling rates was estimated in a range of several to thousands K/my at 773 K. The estimation of the cooling rate from the microstructure in constituent minerals can be applicable to study the thermal history of metamorphic rocks as one of the useful methods.

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