Dependence of exsolution textures in synthetic augite on its composition and cooling rate

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Abstract

Exsolution textures of pyroxene minerals are regarded as effective indicators for the sake of deducing the thermal histories. With the view of elucidating quantitative relations of texture to cooling rate (C.R.) and chemical composition in pyroxenes, the size variations of “spinodally-decomposed (001) lamellae” in synthetic augite were examined with transmission electron microscope (TEM). The textural differences were investigated through cooling rate experiments (cooled over the range 1°C/h to 120°C/h) using homogeneous starting glasses with composition of $\text{Wo}_{32.3}\text{En}_{58.2}\text{Fs}_{9.5}$ and $\text{Wo}_{31.0}\text{En}_{42.7}\text{Fs}_{26.3}$. All experiments were performed under the condition of regulated partial oxygen pressures with a programmed cooling temperature system. Chemical compositions of these samples were determined with electron probe micro-analyzer (EPMA) and analytical electron microscope (AEM). Quantitative relations between lamella wavelength (λ) and C.R. are reported, which were found in the two sets of augites well defined in their composition. The present results include the recalculated data from McCallister (1978) and Grove (1982). The relations between λ and C.R. could be described at the value of $\text{Wo}/(\text{Wo}+\text{En}+\text{Fs})$ fixed to 0.3 as follows:

$$\text{Fs}/(\text{Fs}+\text{En}) = 0 \quad : \quad \lambda = 110 + 400 \times (\text{C.R.})^{-1/3}$$

$$\text{Fs}/(\text{Fs}+\text{En}) = 0.14 \quad : \quad \lambda = 100 + 350 \times (\text{C.R.})^{-1/3}$$

$$\text{Fs}/(\text{Fs}+\text{En}) = 0.40 \quad : \quad \lambda = 70 + 250 \times (\text{C.R.})^{-1/3}$$

(λ in Å and C.R. in °C/h)

The relation of wavelength to cooling rate and composition can be discussed quantitatively and the average cooling rate in natural augite could be appraised by applying these equations.

Introduction

Rock-forming pyroxenes composed of quadrilateral components often show a variety of compositions and characteristic substructures (e.g. exsolution lamellae), and they are considered to be indicators of the thermal history of pyroxenes. There have been commonly recognized two orientations of clinopyroxene lamellae in clinopyroxene host grains with optical microscope (Robinson *et al.*, 1971, 1977; Jaffe *et al.*, 1975). They are nearly parallel to (001) and (100), and are termed (001) lamellae and (100) lamellae, respectively. Significant variables of lattice parameters of the

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host and lamella are a , c and β , which are regarded to be the main determinant of the orientation of lamellae (Robinson *et al.*, 1977). In recent years there have been extensive developments in our understanding of exsolution phenomena through transmission electron microscopic (TEM) observations. The studies of both terrestrial (Dunham *et al.*, 1972; Copley *et al.*, 1974) and lunar (Christie *et al.*, 1971; Lally *et al.*, 1975) pyroxenes implied that differing thermal histories could cause systematic changes in the exsolution textures. Those textures were regarded as the indicator of deducing the cooling history of the exsolved pyroxene. The necessary condition for exsolution to occur is a decrease in free energy on passing from a single phase to a two-phase system. In the process, the exsolution can proceed by two mechanisms; one is a process conventionally known as nucleation and growth and the other is that of the spinodal decomposition (Cahn, 1961). To elucidate the exsolution mechanism, many authors (e.g. McCallister and Yund, 1977; McCallister, 1978; Nord and McCallister, 1979; Grove, 1982; Ried and Fuess, 1986) studied the textural differences through the syntheses of pyroxenes well defined in compositions, with various annealing periods and cooling rates.

McCallister (1978) studied the isothermal coarsening kinetics of (001) exsolution lamellae in synthetic iron-free diopside ($\text{Wo}_{27}\text{En}_{73}\text{Fs}_0$). He measured average lamella wavelengths (λ) as a function of annealing time (t) by TEM and found that his result was consistent with the following relationship derived from the spinodal decomposition:

$$\lambda = \lambda_0 + k(T)t^{1/3} \quad (1)$$

where λ_0 is an initial wavelength of the lamellae, and $k(T)$ is the "kinetic constant" after Park *et al.* (1976). Grove (1982) estimated average cooling rates for the lunar quartz-normative basalt samples from the size variation of exsolution lamellae in augite rims ($\text{Wo}_{30}\text{En}_{42}\text{Fs}_{28}$) (Fig. 1). He proved those mutual relation by the cooling rate experiments (cooled over the range 0.21°C/h to 150°C/h). He used "spinodally-decomposed (001) lamellae" as a cooling rate indicator and defined the cooling rate which controlled the lamella wavelength. Based on the observations of synthetic samples by McCallister (1978) and Grove (1982), we predict that investigations of both thermal and compositional dependences of the lamella wavelength would bring a clear understanding of exsolution mechanism.

The purpose of the present study is to procure a detailed information on deducing the thermal history from the exsolution features in natural pyroxenes. For the above purpose, textural differences in synthetic augite were examined through varying cooling rate and composition.

FIG. 1. The pyroxene quadrilateral showing clinopyroxene compositions in the previous study and those of starting glasses. McCallister and Grove designed isothermal heating experiments and continuous cooling experiments, respectively, and the (001) lamella wavelengths were reported on each sample. The starting glasses were analyzed by EPMA and the analytical points were represented by the solid circles in the figure.

Experimental method

1) Sample characterization

Chemical compositions were analyzed with an automated JEOL model JCSA-733 electron probe micro-analyzer (EPMA) and with a HITACHI model H-600 electron microscope with the attachment of a KEVEX analytical system (AEM). The compositional homogeneity in the samples was insured with EPMA by examining 60 points randomly in the region of 2 mm squares on the samples. Corrections for EPMA were made by the method reported by Bence and Albee (1968). The applied accelerating voltage was 15 kV, and the electron beam diameter was about 1 μm on the specimen throughout the run. Following standard materials were used; Mg: synthetic periclase; Al: synthetic corundum; Fe: natural hematite (Sennin Mine, Iwate, Japan); Si and Ca: synthetic wollastonite. The electron current on specimen was fixed to 0.012 μA on standard periclase. The AEM was used to analyze the bulk compositions in the region of the exsolution textures. Each of the starting glasses well defined in chemical composition (mentioned latter) was used as standard materials for AEM analyses. The corrections were made by the Cliff-Lorimer ratio method (Cliff and Lorimer, 1975).

Thin sections of the run products were prepared for optical microscopic observations. The exsolution textures were observed by a JEOL model 100CX TEM operating at 100 kV equipped with a double-tilt stage. Both ion-thinned and crushed samples from individual cooling rate experiments were examined to measure the wavelength of exsolution lamellae. In order to determine the (001) lamella wavelength, the crystal fragments were tilted that the c^* lay on the horizontal image plate by reference to electron diffraction pattern. The number of lamellae crossing the traverse

on the film was counted, and then the average wavelength was calculated.

2) Sample preparation

Turnock *et al.* (1973) summarized several experimental methods for the syntheses of the quadrilateral pyroxenes at desired compositions. The difficulties encountered in the sample preparations are of controlling oxygen fugacities and of minimizing the chemical interference from the sample container.

The geological importance of the oxygen fugacity control in reactions involving transition elements is emphasized in the literature (e.g. Nafziger *et al.*, 1971). In this study, sample syntheses were all performed in a vertical gas mixing furnace and heating and cooling temperatures were regulated by the programmed controller. The atmosphere within the furnace was maintained at the value near the Fe–FeO equilibrium at given temperature by the reaction of H_2 – CO_2 gas mixture (Nafziger *et al.*, 1971), and the measurement of oxygen fugacity (fO_2) in the furnace was performed by means of solid electrolyte sensor (Sato, 1971). The monitored fO_2 was superimposed on a phase diagram in the system Fe–O (Fig. 2). A Pt.40Rh–Pt.20Rh thermocouple calibrated against the melting point of a gold wire was placed close to the sample at the center of the furnace to monitor the sample temperature in all the way through the heating duration.

FIG. 2. Data for oxygen fugacities in the present experiments. The oxygen fugacities were measured by ZrO_2/Y_2O_3 solid-electrolyte cell under the condition of H_2/CO_2 ratio = 8/7 through all experiments. Data for oxygen fugacities of phase boundaries in the system Fe–O is referred to Nafziger *et al.* (1971).

A platinum crucible is commonly used for the sample container in experimental mineralogy. In this work, iron-bearing starting glasses (sample FFE14 and FFE38) were prepared using a 5 cc platinum crucible, which was treated in advance by heating with the same compositional melts in it so as to impregnate the platinum with iron in order to minimize the iron loss (Turnock *et al.*, 1973). These starting materials weighing about 6 g were compounded from oxide and carbonate mixtures of the desired compositions (Table 1). The initial mixtures were pressed into pellets (6 mm in diameter, 2–3 mm thick and about 500 mg) and heated at 1200°C for 24 hours. The heated pellets turned their color into light brown and then were loaded into the crucible. They were heated again to be completely melted at 1350°C for 10 minutes. The crucible with the charge was took out from the furnace and quenched in the air. The glass thus prepared was ground and remelted to increase the homogeneity. This procedure was repeated three times and the final run product was then used as the starting material for this experiment. The compositions of the starting glasses and their homogeneities were confirmed by EPMA (Fig. 1). The average compositions of the

TABLE 1.

	FEF14	FEF38 (g)
MgO	1.127	0.845
CaCO ₃	1.499	1.499
Fe ₂ O ₃	0.558	1.116
SiO ₂	3.000	3.000
total	6.184	6.460

Chemical compositions of starting mixtures

TABLE 2.

	FEF14	FEF38 (wt%)
MgO	20.14	14.40
CaO	15.59	14.55
FeO	5.87	15.80
SiO ₂	56.60	52.83
Al ₂ O ₃	0.72	0.64
total	98.92	98.22
Ca : Mg : Fe	32.3 : 58.2 : 9.5	31.0 : 42.7 : 26.3

Chemical compositions and molar fractions of Ca, Mg and Fe of starting glasses analyzed by EPMA. Al₂O₃ was added to samples from an Al₂O₃ mortar during grinding. These glasses were used for AEM standards.

starting glasses and their molar fractions of Ca, Mg and Fe are listed in Table 2. These analyses showed that these glass compositions were oversaturated with SiO_2 content in respect of pyroxene stoichiometry.

About 50 mg of the starting glass material was hung on a loop of platinum wire with 0.2 mm in diameter. This method reduces a contact surface of the sample to the holder, and results in a minimum iron loss from the sample and brings a maximum interaction between the sample and gas mixture (Donaldson *et al.*, 1975; Huebner and Turnock, 1980). The starting material was placed in the gas mixing furnace for 3 hours at temperature of 1230°C which is a subsolidus temperature both on FFE14 and FFE38. They were cooled at several constant rates ranging from 1°C/h to 120°C/h and quenched from about 860°C . The quenching method is the same as that for the starting glasses. The run conditions of experiments are shown in Fig. 3 and summarized in Table 3.

FIG. 3. Schematic illustration of experimental conditions. Samples were held at 1230°C for three hours and then cooled at four different rates. The experiments were terminated by quenching in air.

TABLE 3.

cooling rate ($^\circ\text{C/h}$)	Duration (h)	T_{init} ($^\circ\text{C}$)	T_{fin} ($^\circ\text{C}$)
123.2	6	1230	860
26.7	17	1225	865
5.1	70	1230	890
1.1	310	1228	890

Run conditions of cooling rate experiments

Experimental results

1) Chemical compositions of run-products

The compositions of the synthetic augite crystallized from the homogeneous starting glasses were analyzed by EPMA and are plotted in the pyroxene quadrilateral. These plotted points are encircled in Fig. 4. The compositional regions in which exsolution textures were observed by AEM are also shown in Fig. 4. The compositions of Ca, Mg and Fe analyzed by AEM are in good agreement with those by EPMA.

2) Texture observations

Optical microscopic observations indicate that the phases found in these experiments are always clinopyroxene and silica in both samples FFE14 and FFE38. However, the textures produced even at the same cooling rate are quite different between FFE14 and FFE38. The textures of run products of FFE14 and FFE38 from the experiment at 5°C/h in the photomicrographs of thin sections (Fig. 5) show that individual textures produced at the same composition are similar through all cooling rate experiments.

The exsolution textures of the lamellae parallel to (001) were observed under TEM. Fig. 6 shows a series of electron micrographs of the lamellae developed during the cooling experiments. A lamella structure found in the samples heated at 120°C/h and 27°C/h in FFE14 is similar to the modulated exsolution microstructure, which has been observed both in synthetic clinopyroxenes decomposed by isothermal annealing (McCallister, 1978; McCallister and Yund, 1977) and in natural clinopyroxenes decomposed by rapid continuous cooling (Champness and Lorimer, 1971, 1976;

FIG. 4. Compositional variations of exsolved pyroxene. right: EPMA analytical points plotted in the pyroxene quadrilateral were represented by the enclosed areas in the figure. left: AEM analytical points plotted in the quadrilateral represent the bulk compositions of the regions in which exsolution texture were observed.

FIG. 5. Photomicrographs of thin sections showing the textures at 5°C/h experiment on FFE14 (A and B) and FFE38 (C and D).

(A): taken in plane-polarized light. (B): taken under crossed polars. Spherulite texture of pyroxene and SiO_2 intergrowth is observed. (C): taken in plane-polarized light. (D): taken under crossed polars. Porphyritic texture of pyroxene crystals is observed. Crystals of SiO_2 are found as anhedral patches and lath-shaped aggregates. Magnifications of the four photographs are all same and a scale indicator in C represents 1 mm.

Lally *et al.*, 1975). These types of the modulated microstructure are the characteristics of the early stage of spinodal decomposition (Buseck *et al.*, 1980). Therefore, the (001) lamellae were possibly produced by the spinodal mechanism. Lamella wavelengths observed from each micrograph and the average wavelength in each cooling experiment are listed in Table 4. The wavelengths in respect of FFE14 are longer than those of FFE38 through all the cooling rates.

Discussion

Studies of the pyroxene microstructures grown in isothermal annealing play a great role to elucidate the cooling rate (C.R.) by using lamella wavelength (λ). Differentiation of equation (1) yields

FIG. 6. Electron micrographs showing changes in the microstructure with various cooling rates, for synthetic clinopyroxene on FFE14 (A, B, C and D) and FFE38 (E and F). A: 120°C/h, B: 27°C/h, C and E: 5°C/h, D and F: 1°C/h. A scale bar in F indicates 1000 Å and the others also have the same magnification.

TABLE 4.

cooling rate (°C/h)	(001) lamella wavelength measured in augites		
	FEF14	FEF38	(Å)
120	190, 140, 220 [180, 30]*		
27	230, 230, 210 200 [220, 10]	160	
5	290, 340, 290 290, 330, 340 340, 300, 270 [310, 30]	200, 210, 240 230, 230, 200 270 [220, 20]	
1	450, 470, 500 360, 470, 460 480, 420, 370 360 [430, 50]	320, 250, 380 320, 280, 290 240 [300, 50]	

*[mean, σ]

$$d\lambda = k(T)d(t^{1/3}). \quad (2)$$

In case of linear cooling, temperature (T) and time (t) obey the following equation:

$$T = T_i - (C.R.)t \quad (3)$$

where T_i is the initial temperature of the exsolution process. Substitution of equation (3) to (2) and integration of both sides yields

$$\int_{\lambda_o}^{\lambda} d\lambda = \int_0^{\Delta t} k(T_i - (C.R.)t) d(t^{1/3}) \quad (4)$$

where λ_o is the initial lamella wavelength and Δt is the time range over which lamellae developed. In the period of Δt , exsolution occurs over the temperature range T_i to $T_i - (C.R.) \Delta t$. The Δt can be calculated by the final temperature (T_f); $T_i - (C.R.) \Delta t$ as follows:

$$\Delta t = (T_i - T_f)/C.R. \quad (5)$$

Substitution of equation (5) to (4) and replacement of (C.R.) t to x^3 in the right side yields

$$\int_{\lambda_o}^{\lambda} d\lambda = (C.R.)^{-1/3} \int_0^{(T_i - T_f)^{1/3}} k(T_i - x^3) dx. \quad (6)$$

If four parameters of λ_o , $k(T)$, T_i and T_f are known, then the lamella wavelength for the corresponding cooling rate will be calculated from equation (6). This equation can be simplified to the form

$$\lambda = \lambda_o + C(T_i, T_f, k(T)) \times (C.R.)^{-1/3} \quad (7)$$

where $C(T_i, T_f, k(T))$ is a constant.

McCallister (1978) found that the coarsening kinetics of the synthetic clinopyroxene ($Wo_{27}En_{73}Fs_0$) obeys the $t^{1/3}$ law in isothermal annealing procedure. The following rate constant was derived on the basis of the Fig. 3 in McCallister (1978):

$$\ln k(T) = -17.3 \times 1000/T + 16.3 \quad (T \text{ in K}). \quad (8)$$

He assumed that the coherent spinodal in that composition was at approximately 1335°C, because a modulated exsolution microstructure was developed at 1325°C, but was not found at 1345°C (McCallister and Yund, 1977). The form of the kinetic constant ($k(T)$) predicts that the decomposition does not occur at relatively low temperatures. The lamella wavelength calculated over the temperature range 1335°C–1000°C from the equation (6) explains that the mobility of the species, which effectively controls the overall rate of coarsening, decreases and that the lamella coarsening may almost cease at temperature below 1000°C. Thus, the coarsening may occur at temperatures between 1335°C and about 1000°C for the particular composition used in his investigation. In general, when the augite is cooled from T_i to a specific temperature, below which exsolution does not occur, the size for lamella wavelength can be calculated from equation (6) by three parameters of λ_o , $k(T)$ and T_i . The lamella wavelength for the corresponding cooling rate is now calculated by equation (6) as follows, assuming that T_i is 1335°C, T_f is 1000°C and λ_o is 110 Å, which is derived from his result:

$$\lambda = 110 + 400 \times (C.R.)^{-1/3} \quad (\text{Å}) \quad (9)$$

where C.R. is given in °C/h.

The equation (9) is shown as an upper straight line in the graph with wavelength vs $(C.R.)^{-1/3}$ (Fig. 7). If the wavelength is known, the cooling rate over the temperature range about 1300°C–1000°C can be deduced from the equation (9).

Grove (1982) examined the effects of cooling rate on the size of lamella wavelength in augite rims ($Wo_{30}En_{42}Fs_{28}$) in the linear cooling rate experiment. He showed that the lamella coarsening almost ceased below 800°C by integrating the rate constant proposed by him. He presented the size of “spinodally-decomposed lamellae” on a

FIG. 7. Lamella wavelength (λ)—cooling rate—composition diagram. The open circle and the solid circle in the figure represent the mean lamella wavelengths measured in augites on FFE14 and FFE38, respectively.

$t^{1/3}$ plot and described the following equation by the rate law, assuming that the most of the lamella coarsening occurred over the temperature range from 1100°C to 800°C:

$$\lambda = 70 + 107.3 \times (\Delta t)^{1/3} \quad (\text{\AA}) \quad (10)$$

where Δt is the time occupied on the lamella coarsening from 1100°C to 800°C given in days.

The linear C.R. is expressed from equation (5) as follows:

$$\text{C.R.} = (T_i - T_f) / \Delta t \quad (11)$$

Substitution of equation (11) to equation (7) yields

$$\lambda = \lambda_0 + C(T_i, T_f, k(T))(T_i - T_f)^{-1/3} \times (\Delta t)^{1/3} \quad (12)$$

The equation (12) provides the identical lamella wavelength with the equation (10) which was experimentally determined, subject to the conditions of $T_i=1100^\circ\text{C}$ and $T_f=800^\circ\text{C}$. The relations between λ and C.R. can be described as the same form with the equation (7) as follows:

$$\lambda = 70 + 250 \times (\text{C.R.})^{-1/3} \quad (\text{\AA}) \quad (13)$$

The relation between lamella wavelength and cooling rate for temperature range $1100^\circ\text{C}-800^\circ\text{C}$ is shown as a lower straight line in Fig. 7.

The present experimental data on FFE38 are plotted in Fig. 7. These results are in well accord with the recalculated results from the studies of Grove (equation (13)). The experimental data on FFE14 is plotted in Fig. 7. The regression analysis (a middle line in the graph) of the data on 27°C/h , 5°C/h and 1°C/h experiments gives a following equation, assuming that λ and C.R. have the same relation with those of equation (7) and λ_0 is $100 \text{ \AA}$:

$$\lambda = 100 + 350 \times (\text{C.R.})^{-1/3} \quad (\text{\AA}) \quad (14)$$

The dependence of wavelength on cooling rate and composition can be discussed quantitatively and the average cooling rate in natural augite ($W_o/(W_o+En+Fs)=0.3$, $Fs/(Fs+En)=0\sim 0.4$) could be appraised by applying the wavelength-cooling rate-composition diagram and/or equation (9), (13) and (14).

A small compositional changes in components of En and Fs can lead a shift of the temperature of coherent spinodals in the pseudobinary joins. Traversing across the pyroxene quadrilateral toward the En-Di join, the proposed coherent spinodals are shifted to the higher temperature (see Fig. 1, Jantzen, 1984). The shifting of the spinodals is consistent with the behavior of the lamella coarsening reported in this paper, which occurs at about 1300°C in the case for $Fs/(Fs+En)=0$ and about 1100°C for $Fs/(Fs+En)=0.4$. At temperatures below about 1000°C for $Fs/(Fs+En)=0$ and below about 800°C for $Fs/(Fs+En)=0.4$, respectively, the coarsening is inhibited by decreasing the mobility of the rate-controlling species. In summary, the temperature range producing lamella coarsening is about $1300^\circ\text{C}-1000^\circ\text{C}$ for $Fs/(Fs+En)=0$ and about $1100^\circ\text{C}-800^\circ\text{C}$ for $Fs/(Fs+En)=0.4$. The temperature differences in the respective cases are both almost 300°C , and they may be regarded to be constant regardless of the compositions through $Fs/(Fs+En)=0$ to $Fs/(Fs+En)=0.4$. On occasion for deducing the thermal histories of natural augites by using exsolution lamellae, it should be noted that the temperature range over which the exsolution proceeded

shifts corresponding to the Fs/En ratio.

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