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ORIENTED EXSOLUTION OF AUGITE IN PIGEONITE

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Abstract

Pigeonite crystals from the Moore County meteorite show oriented exsolutions of two augites parallel to (001) and (100). The host and the exsolved phases indicate two-dimensional agreement not only in orientation but also in cell dimensions along their boundary planes. On the assumption that the two augites have an identical chemical composition and the difference between the two augites in the cell dimensions is due to lattice distortions in the exsolution, the cell dimensions corrected for the lattice distortions were estimated by using the elastic constants. The strain energies were estimated to be 0.02 and 0.04 kcal/mole for augites exsolved parallel to (001) and (100) respectively on the basis of the changes of the cell dimensions.

Pigeonite crystals from the Skaergaard intrusion were also studied for comparison. The exsolved augite is observed only in one orientation, which is not perfectly parallel to (001) of the pigeonite, and shows its intrinsic cell dimensions free from lattice distortions. This strongly suggests that pigeonite from the Moore County meteorite represents an intermediate stage in the process of exsolution of augite.

Introduction

The most important pyroxenes in igneous rocks belong to the quadrilateral composition area of the $CaMgSi_2O_6$-$CaFeSi_2O_6$-$MgSiO_3$-$FeSiO_3$ system. Pyroxenes of this quadrilateral area are divided into two groups: Ca-rich and Ca-poor pyroxenes. They usually show various types of exsolution of one from the other, developed at subsolidus temperatures. The exsolution phenomena in pyroxenes have received much attention because they provide a great potential for storing information on the phase relations and the genetic relations of the pyroxenes involved as well as the rocks containing them.

Hess (1941) and Poldervaart and Hess (1951) studied the exsolution phenomena of pyroxenes in plutonic igneous rocks by means of the optical method and interpreted them on the basis of the phase relations of the $MgSiO_3$-$FeSiO_3$ system (Bowen & Schairer, 1935). Their interpretations have been widely accepted since then. Bown and Gay (1959, 1960) studied the exsolution phenomena in pyroxenes, especially in pyroxenes from the Skaergaard intrusion, by means of the X-ray single crystal method. Most of their results confirmed those obtained by the optical method (Brown, 1957) and were explained by the ordinary accepted interpretation.

However, Sclar *et al.* (1964) reported a new phase relation of $MgSiO_3$, in which clinoenstatite was stable at lower temperatures than orthoenstatite under high pressures. Similar relations were reported with $FeSiO_3$ by Lindsley *et al.* (1965) and Akimoto *et al.* (1965). Since then, the phase relations of the Ca-poor pyroxenes as well as those between the Ca-rich and Ca-poor pyroxenes have been widely studied, though they are still unsolved. In order to reexamine the exsolution phenomena in pyroxenes on the basis of the present knowledges of the phase relations of the related pyroxenes, many crystals of pigeonite and augite from varied environment have been studied by the X-ray single crystal method (Morimoto, 1966, and in preparation).

Among the crystals examined, pigeonite crystals from the Moore County, North Carolina, meteorite, given to one of us (N. M.) by Dr. H. S. Yoder, Jr., Geophysical Laboratory, Washington, D. C., were found to have oriented exsolutions of augites not only parallel to (001) but also (100). Because the exsolution of augite parallel to (100) in pigeonite was found for the first time, the two oriented exsolutions of augites in the pigeonite crystals were studied in detail in this investigation.

SPECIMEN DESCRIPTION

The pyroxenes in the Moore County meteorite were first described by Henderson and Davis (1936) and later by Hess and Henderson (1948) in more detail. Hess and Henderson confirmed four phases of pyroxenes: pigeonite, augite, hypersthene and salite. They described their observation on the thin section of the Moore County meteorite as follows:

"The bulk of the pyroxene present is pigeonite. It has beautifully developed augite lamellae parallel to {001}." "Small roughly quadrilateral areas bounded by cleavages and augite lamellae have inverted to hypersthene in a few of the pigeonite crystals." "This hypersthene . . . has within it the very fine exsolution lamellae of clinopyroxene parallel to the {100} plane of the orthopyroxene . . . ."

They estimated the composition of large metal atoms in the original pigeonite to be $Mg_{46}Fe_{44}Ca_{10}$ on the basis of the chemical analysis. The compositions of pigeonite and exsolved augite they observed in the thin section were estimated to be $Mg_{47.5}Fe_{47.5}Ca_5$ and $Mg_{36}Fe_{21}Ca_{43}$ respectively based on the optical properties. The compositions of hypersthene and the exsolved clinopyroxene (salite) in it were also estimated in the same way to be $Mg_{49.5}Fe_{50.5}$ and $Mg_{32}Fe_{18}Ca_{50}$ respectively.

Dr. F. Boyd, Geophysical Laboratory, Washington, D. C., examined pigeonite crystals of the Moore county meteorite by the electron-probe microanalyser and gave the composition of $Mg_{48.14}Fe_{46.24}Ca_{5.62}$ for the pigeonite phase. According to him, the variation of the chemical composition in the pigeonite phase is greater even in one crystal compared with pigeonite crystals from the Isles of Mull (Morimoto and Güven, 1968). The values given above are the average of results obtained on about 20 points in ten grains. Calcium counts varied ±10 percent from the mean given above.

EXPERIMENTS

Apparent single crystals of pigeonite and hypersthene, examined under the microscope, were studied by the precession and Weissenberg methods with MoKα(0.7107 Å) and CuKα(1.5418 Å) radiations.

The X-ray photographs of the pigeonite crystals show that there are two or three different phases besides the main pigeonite phase. They are hypersthene and two kinds of augite, and their orientations relative to pigeonite are always definite. Although hypersthene is often missing, one or two kinds of augite are always present. The precession photographs (Fig. 1 and Fig. 2 a) show the relative orientations of two kinds of augite and pigeonite. All three phases share the *b* axis in common. One of the augites shares (001), and the other (100) with pigeonite. They are called augite (c) and augite (a) respectively in this investigation. When hypersthene appears together with other phases, it has the *b* axis in common with all other phases and shares (100) with pigeonite (Fig. 2 b).

Examination of several pigeonite crystals by the X-ray single-crystal method (Table 1) shows most of each crystal consists of pigeonite, and the other phases are small in amount

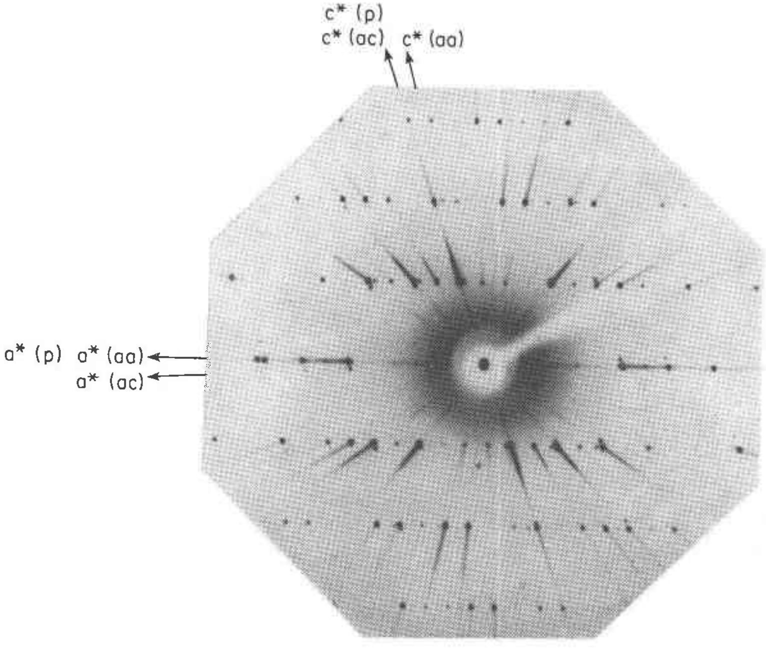

FIG. 1. Precession photograph $(010)_0^*$ of pigeonite from the Moore County meteorite with two exsolved augites. Mo Kα (Zr-filtered), $\beta = 30°$, 20 hours. The a^* and c^* axes of pigeonite, augite (c) and augite (a) are indicated. a^*(p), c^*(p): pigeonite; a^*(ac), c^*(ac): augite (c); a^*(aa), c^*(aa): augite (a).

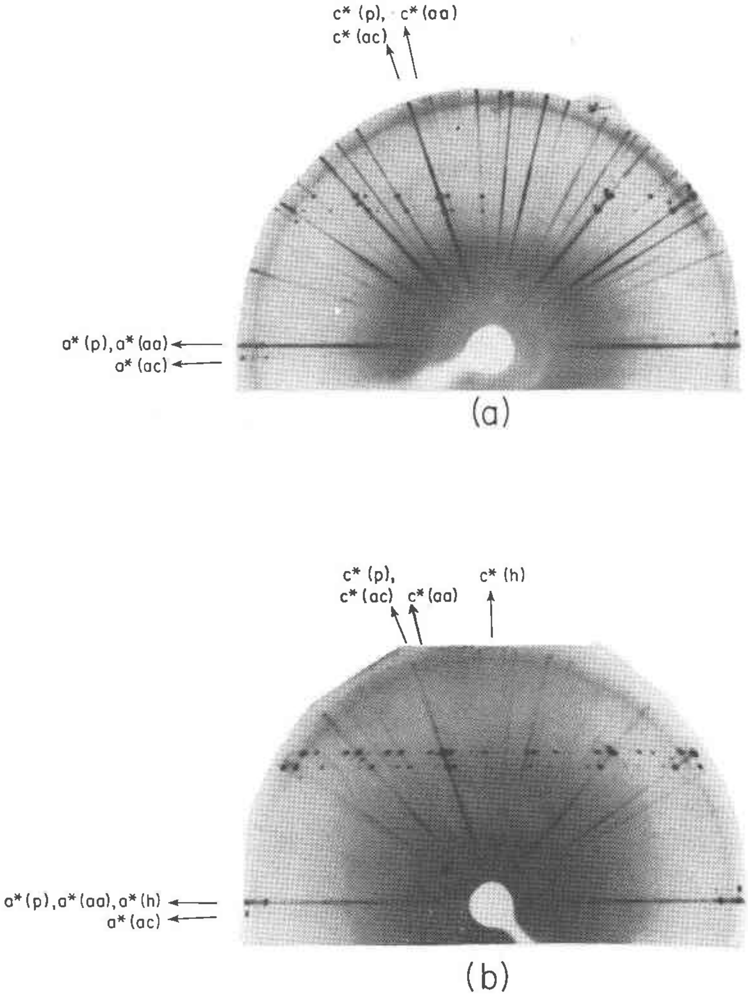

FIG. 2. Precession photograph $(010)_0^*$ of pigeonites from the Moore County meteorite with two exsolved augites (a) and with hypersthene and two exsolved augites (b). Cu $K\alpha$, $\mu=30°$, 10 hours. The a^* and c^* axes of pigeonite, two exsolved augites and hypersthene are indicated. a^*(p), c^*(p) : pigeonite; a^*(ac), c^*(ac) : augite (c); a^*(aa), c^*(aa) : augite (a); a^*(h), c^*(h) : hypersthene.

compared with pigeonite. Though augite (c) is always greater than augite (a) in amount, their ratio is variable. The amount of hypersthene is quite variable from crystal to crystal. Augite (a) appears without any hypersthene and is almost equal to augite (c) in amount in some pigeonite crystals. The X-ray reflections from hypersthene in pigeonite crystals are always slightly diffuse with streaks along the a^* axis, though the reflections from the other phases are sharp. Hypersthene crystals with practically no pigeonite phase were also found.

TABLE 1. COEXISTING PHASES IN PIGEONITE AND HYPERSTHENE CRYSTALS FROM THE MOORE COUNTY METEORITE CONFIRMED BY THE X-RAY SINGLE-CRYSTAL METHOD[a]

Crystals	Host	Augite (c)	Augite (a)
Pigeonite No. 1	p(h)	o	o
Pigeonite No. 2	p	o	o
Pigeonite No. 3	p(h)	o	o
Hypersthene No. 4	h	—	o
Pigeonite No. 5	p	o	o
Pigeonite No. 6	p	o	—
Pigeonite No. 7 (heated)	p(h)	o	—
Hypersthene No. 8 (heated)	h	—	o
Pigeonite No. 9 (heated)	p	o	—

[a] p: pigeonite. h: hypersthene. p(h): pigeonite with hypersthene. o: observed. —: not observed.

They have small amount of augite sharing (100) in common. The X-ray reflections from both phases are sharp. The results on the pigeonite and hypersthene crystals (Table 1) indicate that augite (a) is different from salite F described by Hess and Henderson (1948), which was exsolved in hypersthene parallel to its (100) and was considered to occupy only one percent of the whole volume of pyroxenes.

The cell dimensions of pigeonite were obtained by the least-squares method by Burnham's (1965) program using 40 independent reflections measured on the precision back-reflection Weissenberg photographs. The dimensions of the exsolved augites and hypersthene were obtained by the precession photographs calibrated by the host pigeonite. The dimensions of augite (c) and augite (a) are given together with those of pigeonite and hypersthene (Table 2). Pigeonite and augite (c), sharing (001), have the same orientation and equal length of *a* and *b*. A shrinkage of *a* in augite (c), seems to be compensated by an expansion of *c*. Pigeonite and augite (a), sharing (100), have the same orientation and equal length of *b* and *c*, resulting in an expansion of *a* of augite (a) to compensate a shrinkage of *c*. These observations on the orientations and the cell dimensions of pigeonite and augites in the pigeonite crystals indicate that exsolutions of augites took place in pigeonite parallel to (001) or (100) sharing (001) or (100), respectively. Thus the augites underwent readjustments in the direction normal to the boundary plane to take account of the change in area at the boundary plane. The X-ray reflections from the exsolved augites are sharp indicating that their cell dimensions are constant over their whole volume.

When hypersthene exists in the pigeonite crystals, having the common *b* and *c* axes with other phases, its *a* is equal to $2a \sin\beta$ of pigeonite in the limit of experimental error (Table 2). Thus the cell volume of the hypersthene is exactly twice greater than that of pigeonite.

In order to examine the effect of heating on the exsolved augites, pigeonite and hypersthene crystals from the Moore County meteorite were heated at 850°C for 48 hours. They were enclosed in an evacuated silica tube with pure iron grains. The tube was quenched to room temperature after heating. Three crystals in the quenched products were examined by the X rays. No augite (a) was observed in two pigeonite crystals (Table 1). Because some crystals (No. 6 in Table 1) have exsolution of only augite (c) before heating and only two heated crystals of pigeonite were examined, these results do not necessarily mean that

TABLE 2. CELL DIMENSIONS AND CHEMICAL COMPOSITIONS OF PIGEONITE, HYPERSTHENE AND TWO EXSOLVED AUGITES, AUGITE (c) AND AUGITE (a), COEXISTING IN PIGEONITE CRYSTALS FROM THE MOORE COUNTY METEORITE. THE CELL DIMENSIONS OF AUGITE (f) ARE OBTAINED FROM THOSE OF THE TWO EXSOLVED AUGITES BY THE METHOD DESCRIBED IN TEXT

Pyroxenes	a(Å)	b(Å)	c(Å)	β( )	V(Å^3)	Composition
pigeonite (host)	9.690 (1)	8.942 (1)	5.227 (1)	108.752 (6)	428.70 (15)	$Mg_{47.5}Fe_{47.5}Ca_{5.0}$ (Hess & Henderson, 1948) $Mg_{48.2}Fe_{46.2}Ca_{5.6}$ (Boyd, priv. comm.)
hypersthene	18.35 (1)	8.94 (1)	5.23 (1)	—	859.4	$Mg_{47.5}Fe_{47.5}Ca_{5.0}$ (Hess & Henderson, 1948)
augite (c)	9.69 (1)	8.94 (1)	5.28 (1)	106.0 (1)	439.7	$Mg_{36}Fe_{21}Ca_{43}$ (Hess & Henderson, 1948)
augite (a)	9.77 (1)	8.94 (1)	5.23 (1)	105.9 (1)	439.3	
augite (f)	9.74	8.94	5.27	106.0	441.1	$Mg_{38}Fe_{19}Ca_{43}$

augite (a) disappeared in the pigeonites by the heating. No change of the cell dimensions of pigeonite were observed to within 0.005 Å by heating.

CHANGE OF CELL DIMENSIONS OF AUGITE DUE TO EXSOLUTION

Because it seemed unlikely that the strain at the exsolution boundary could control the cell dimensions of the exsolved phases over many hundreds of unit cells regardless of their chemical composition, the two augites were, at first, considered to have been exsolved at different stages of cooling and have different chemical compositions (Morimoto, 1966). Professor J. V. Smith, however, pointed out equality of the cell volumes of the two exsolved augites (Table 2) and suggested possibility of their indentical chemical composition on the basis of his study on perthites (Smith, 1961).

In the following, the exsolution of augites in the Moore County pigeonite has been studied on the assumption that augite (c) and augite (a) have an identical chemical composition and the difference between their cell dimensions is due to lattice distortions by orientation effects. Propriety of this assumption will be discussed later.

We shall first assume that an augite becomes augite (c) or augite (a) when the augite is subjected to forced deformation parallel to the boundary plane with pigeonite and to free deformation in direction normal to the plane so as to reduce its strain energy to a minimum. The augite, free from any deformation, is called augite (f) in the present investigation.

A component, ϵ_{ij}, of a strain tensor in the Cartesian coordinates, O-$x_1x_2x_3$, is defined by the variation of the displacement u_i with x_i as follows,

$$\epsilon_{ij} = \frac{1}{2}\left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i}\right) \tag{1}$$

where the subscripts i and j run from 1 to 3. A component, ϵ_i, in the matrix expressing the strain is given by the component of the tensor as follows, $\epsilon_1=\epsilon_{11}$, $\epsilon_2=\epsilon_{22}$, $\epsilon_3=\epsilon_{33}$, $\epsilon_4=2\epsilon_{23}$, $\epsilon_5=2\epsilon_{13}$ and $\epsilon_6=2\epsilon_{12}$. When the strain is small, the stress, σ_i, is expressed as follows,

$$\sigma_i = \sum_j c_{ij}\epsilon_j, \tag{2}$$

where c_{ij} is the elastic constant or stiffness, and i and j run from 1 to 6. Then the strain energy, E, becomes (Nye, 1957),

$$E = \frac{1}{2}\sum_i \sigma_i\epsilon_i = \frac{1}{2}\sum_{ij} c_{ij}\epsilon_i\epsilon_j \tag{3}$$

From this equation, we obtain,

$$\frac{\partial E}{\partial \epsilon_i} = \sum_j c_{ij}\epsilon_j \tag{4}$$

When topotatic relations exist as in the Moore County pigeonite, the strains in the exsolved phases are considered to be small, and the relations mentioned above can be used to estimate the strains.

When a monocline lattice $M(a, b, c, \beta)$ deforms to $M'(a', b', c', \beta')$, the components of the strain tensor are given as follows by the definition of equation (1).

$$\epsilon_{11} = \frac{a'\sin\beta' - a\sin\beta}{a\sin\beta}$$

$$\epsilon_{22} = \frac{b' - b}{b}$$

$$\epsilon_{33} = \frac{c' - c}{c}$$

$$\epsilon_{23} = 0$$

$$\epsilon_{13} = \frac{1}{2}\left(\frac{a'\cos\beta'}{a\sin\beta} - \frac{c'\cos\beta}{c\sin\beta}\right)$$

$$\epsilon_{12} = 0$$

where b and c are taken parallel to the X_2 and X_3 axes, respectively. The change of ϵ_{ij} in the deformation from augite (c) to augite (a), that is, ϵ_{ij} (c→a), can be calculated by substituting the values in Table 2, into the above relations,

$$\epsilon_{11}(c \rightarrow a) = 0.0087 \tag{5}$$

$$\epsilon_{22}(c \rightarrow a) = 0 \tag{6}$$

$$\epsilon_{33}(c \rightarrow a) = -0.0095 \tag{7}$$

$$\epsilon_{13}(c \rightarrow a) = -0.0017 \tag{8}$$

Because augite (f) is to appear on the way from augite (c) to augite (a), the following equation is easily obtained,

$$\begin{aligned} \epsilon_{ij}(c \rightarrow a) &= \epsilon_{ij}(c \rightarrow f) + \epsilon_{ij}(f \rightarrow a) \\ &= -\epsilon_{ij}(c) + \epsilon_{ij}(a) \end{aligned} \tag{9}$$

The equation (6) indicates that ϵ_{22} (a) -ϵ_{22} (c) = 0. Possible errors for the values obtained above are 0.002 for ϵ_{11} and ϵ_{22}, 0.004 for ϵ_{33} and 0.003 for ϵ_{13}. Thus, the values of -0.0017 in equation (8) can be considered to be zero within experimental error. Furthermore, augite (c) and augite (a) have the identical values of b and β, which are observed in free augite crystals with the composition range corresponding to the exsolved augites. It is, therefore, reasonable to assume that neither strain b nor shear stress across the lamellae plane took place in the deformation from augite (c) to augite (a). Thus the contributions of ϵ_{13} to the strain energies for augite (a) and augite (c) are considered to be very small. In the following calculation, we assume that ϵ_{13} can be omitted without changing the basic nature of the results (see Appendix). The variables to be determined are,

$$\epsilon_{11}(\mathrm{a}) = \overset{\mathrm{a}}{\epsilon}_1, \quad \epsilon_{33}(\mathrm{a}) = \overset{\mathrm{a}}{\epsilon}_3, \quad \epsilon_{11}(\mathrm{c}) = \overset{\mathrm{c}}{\epsilon}_1 \quad \text{and} \quad \epsilon_{33}(\mathrm{c}) = \overset{\mathrm{c}}{\epsilon}_3.$$

Among them, $\epsilon_3{}^{\mathrm{a}}$ and $\epsilon_1{}^{\mathrm{c}}$ are caused by forced deformation and $\epsilon_1{}^{\mathrm{a}}$ and $\epsilon_3{}^{\mathrm{c}}$ are by free deformation to reduce the strain energy of each exsolved augite. For a minimum strain energy the equation (4) must be zero with regard to the component of free deformation. Substituting the non-zero components into the equation (4), we obtain,

$$c_{13}\overset{\mathrm{c}}{\epsilon}_1 + c_{33}\overset{\mathrm{c}}{\epsilon}_3 = 0, \tag{10}$$

$$c_{11}\overset{\mathrm{a}}{\epsilon}_1 + c_{13}\overset{\mathrm{a}}{\epsilon}_3 = 0. \tag{11}$$

From the equation (9), the following relations are obtained,

$$-\overset{\mathrm{c}}{\epsilon}_1 + \overset{\mathrm{a}}{\epsilon}_1 = \epsilon_{11}(\mathrm{c} \rightarrow \mathrm{a}) \tag{12}$$

$$-\overset{\mathrm{c}}{\epsilon}_3 + \overset{\mathrm{a}}{\epsilon}_3 = \epsilon_{33}(\mathrm{c} \rightarrow \mathrm{a}) \tag{13}$$

Aleksandrov *et al.* (1964) gave the following values of c_{ij}'s for augite ($\times 10^{11}$ dyne/cm^2): $c_{11}=18.16$, $c_{22}=15.07$, $c_{33}=21.78$, $c_{44}=6.97$, $c_{55}=5.11$, $c_{66}=5.58$, $c_{12}=7.34$, $c_{13}=7.24$, $c_{23}=3.39$, $c_{15}=1.99$, $c_{25}=1.66$, $c_{35}=2.46$ and $c_{46}=0.43$. By substituting these values and the values of equations (5) and (7) into equations (10), (11), (12), and (13), we have,

$$\overset{c}{\epsilon_1} = -\,0.0057$$

$$\overset{c}{\epsilon_3} = 0.0019$$

$$\overset{a}{\epsilon_1} = 0.0030$$

$$\overset{a}{\epsilon_3} = -\,0.0076.$$

In conclusion, *a* contracts 0.6 percent for augite (c) and expands 0.3 percent for augite (a), while *c* expands 0.2 percent for augite (c) and contracts 0.8 percent for augite (a). These relations give the cell dimensions of augite (f) (Table 2).

STRAIN ENERGY OF THE EXSOLVED PHASES

Based on the cell dimensions of the free augite (f) and those of the exsolved augite (c) and augite (a), it is possible to estimate the strain energies of augite (c), E^c, and of augite (a), E^a, from the equation (3). They are,

$$E^c = 2.6 \times 10^7 \text{ erg/cm}^3,$$

$$E^a = 5.5 \times 10^7 \text{ erg/cm}^3.$$

Using the molar volume of 33 cm^3 for each augite, we obtain,

$$E^c = 0.02 \text{ kcal/mole},$$

$$E^a = 0.04 \text{ kcal/mole}.$$

In the similar way, we can estimate the strain energies of augites exsolved parallel to (010) or cleavage plane $\{110\}$ of pigeonite, on the condition that the host and the exsolved phases retain two-dimensional agreement in their cell dimensions along the boundary planes. If pigeonite exsolves augite on (010), we have,

$$\overset{b}{\epsilon_1} = \overset{b}{\epsilon_{11}} = -\,0.0048$$

$$\overset{b}{\epsilon_3} = \overset{b}{\epsilon_{33}} = -\,0.0076$$

$$\overset{b}{\epsilon_5} = 2\overset{b}{\epsilon_{13}} = +\,0.0473.$$

Because a free deformation takes place perpendicular to (010) so that the strain energy E^b becomes minimum, we have the following relation from the equation (4),

$$\frac{\partial E^{\mathrm{b}}}{\partial \epsilon_2^{\mathrm{b}}} = c_{12}\epsilon_1^{\mathrm{b}} + c_{22}\epsilon_2^{\mathrm{b}} + c_{23}\epsilon_3^{\mathrm{b}} + c_{25}\epsilon_5^{\mathrm{b}} = 0.$$

This gives $\epsilon_2^{\mathrm{b}} = -0.0012$, and $E^{\mathrm{b}} = 55 \times 10^7$ erg/cm$_3$ = 0.43 kcal/mole. For the exsolution on $\{110\}$, the components of the strain tensor, ϵ_{12} and ϵ_{23}, are not zero and the lattice of augite can not be monoclinic. We obtain the strain energy E^{m} of about 0.3 kcal/mole in very much the same way.

So far we have discussed the strain energy of augites exsolved in pigeonite on the condition that no stress applied on the pigeonite. When the pigeonite was under a hydrostatic pressure, the discussions mentioned above can be applied without changing the results. However, if the pigeonite exsolves augites under a stress and has a strain represented by ϵ_i^{p}, we have to consider the effect of an additional stress on the augites to evaluate their strain energies. We shall here find what relations are necessary among ϵ_i^{p}'s for the following condition,

$$E^{\mathrm{a}} = E^{\mathrm{c}}. \tag{14}$$

As discussed above, if neither strain along *b* nor shear stress across the lamellae plane takes place and monoclinic symmetry is retained in deformation from augite (f) to augite (c) or to augite (a), ϵ_i^{a} and ϵ_i^{c} are considered to be zero for $i = 2$, 4, 5 and 6. The contributions of ϵ_i^{p}'s, with $i = 2$, 4, 5 and 6, to E^{a} and E^{c} are small compared with those of ϵ_1^{p} and ϵ_3^{p} and can be neglected. Then the equation (14) becomes,

$$\begin{aligned} &\tfrac{1}{2}c_{11}(\epsilon_1^{\mathrm{a}} + \epsilon_1^{\mathrm{p}})^2 + \tfrac{1}{2}c_{33}(\epsilon_3^{\mathrm{a}} + \epsilon_3^{\mathrm{p}})^2 + c_{13}(\epsilon_1^{\mathrm{a}} + \epsilon_1^{\mathrm{p}})(\epsilon_3^{\mathrm{a}} + \epsilon_3^{\mathrm{p}}) \\ &= \tfrac{1}{2}c_{11}(\epsilon_1^{\mathrm{c}} + \epsilon_1^{\mathrm{p}})^2 + \tfrac{1}{2}c_{33}(\epsilon_3^{\mathrm{c}} + \epsilon_3^{\mathrm{p}})^2 + c_{13}(\epsilon_1^{\mathrm{c}} + \epsilon_1^{\mathrm{p}})(\epsilon_3^{\mathrm{c}} + \epsilon_3^{\mathrm{p}}). \end{aligned}$$

By substituting the known values of ϵ_1^{a}, ϵ_3^{a}, ϵ_1^{c} and ϵ_3^{c} and the values of c_{ij}'s for augite into the above equation, we have,

$$\epsilon_1^{\mathrm{p}} = 1.6\epsilon_3^{\mathrm{p}} - 0.0042. \tag{15}$$

If we substitute this relation into equation (2), we obtain,

$$\sigma_1^{\mathrm{p}} = 1.1\sigma_3^{\mathrm{p}} - 4.3 \times 10^9 \ \mathrm{dyne/cm^2}. \tag{16}$$

Equation (16) means that the relation (14) is satisfied when an additional strain corresponding to an additional pressure of about 4,000 bars applies on (100) of pigeonite in comparison with the strain on its (001).

DISCUSSION

Two augites exsolved in different orientations are usually observed in *inverted pigeonites*. Poldervaart and Hess (1951) considered them as products at different stages of cooling with different compositions, and gave the following interpretation: the crystallization of the original pigeonite was followed, upon cooling, by exsolution of augite plates parallel to (001); by the further cooling, the pigeonite inverted to hypersthene, which exsolved fine lamellae of another augite parallel to (100). However, the study on exsolutions in *pigeonites* from various terrestrial rocks gives the following results: two augites in two different orientations, approximately parallel to (001) and (100), are found in pigeonites from volcanic rocks and are considered to have been exsolved at the same stage of cooling; pigeonites from plutonic rocks show exsolution of augite only parallel to or approximately parallel to (001) (Morimoto, in preparation). Thus it is very probable that two augites were simultaneously exsolved in pigeonite from the Moore County meteorite and the difference between their compositions are very slight, if any. The discussion given below also supports this conclusion.

COMPOSITIONS AND CELL DIMENSIONS OF EXSOLVED AUGITES

The direct application of the observed values of $a\ \sin\beta$ and b of augite (c) and augite (a) to the diagrams by Brown (1960) and Viswanathan (1966) gives the compositions $Mg_{24}Fe_{44}Ca_{32}$ and $Mg_{17}Fe_{35}Ca_{48}$ to augite (c) and augite (a) respectively. Hess and Henderson (1948), however, estimated the composition $Mg_{36}Fe_{21}Ca_{43}$ for augite (c) on the basis of the optical properties. If the values of $a\ \sin\beta$ and b estimated for augite (f) (Table 2) are used in the diagrams mentioned above, we obtain the composition $Mg_{38}Fe_{19}Ca_{43}$ which is in remarkable agreement with the composition of augite (c) by Hess and Henderson. Thus it seems certain that the exsolved augites retain lattice distortions by orientation effects in pigeonite and the cell dimensions corrected for the lattice distortions represent their composition satisfactorily.

Smith (1961) explained the orientation effects in perthites semiquantitatively by assuming that the volume of a perthite unit remains constant even if when the lattice is distorted. Based on this assumption, he estimated the lattice distortions and obtained the probable compositions of perthite units. In this investigation, we have been able to observe the cell dimensions of the exsolved augites in two different orientations in pigeonite. This made it possible to estimate the corrected cell dimensions for the orientation effects by using the elastic constants and to obtain the strain energies without assuming a constant cell volume for the exsolved components.

The procedures given in this study can be applied even if when the two exsolved components have slightly different compositions, by adding parameters which can represent the relationship between the compositions of the two components.

EXSOLVED AUGITE IN PIGEONITE FROM THE SKAERGAARD INTRUSION

After the effects of lattice distortions on the cell dimensions had been estimated for the exsolved augites in pigeonite from the Moore County meteorite, we had a chance to examine pigeonite from the Skaergaard intrusion, which was given by Professor G. M. Brown. Pigeonite crystals from ferrogabbro 4430 of the intrusion, with the chemical composition $Mg_{45.5}Fe_{45.7}Ca_{8.6}$ (Brown, 1957), were studied by the Weissenberg and precession methods. The crystals contain hypersthene and exsolved augite and all the three phases share a common *b* axis. The (001) plane of the augite is almost, but not quite, parallel to that of the pigeonite: the two planes make 0.9±0.1°. This relationship is clearly observed in the precession photograph $(010)^*_0$(Fig. 3) in comparison with Figure 1. Since the cell dimensions of the pigeonite and hypersthene (Table 3) obtained from the precession photographs, are in complete agreement with those of pigeonite and hypersthene from the Moore County meteorite (Table

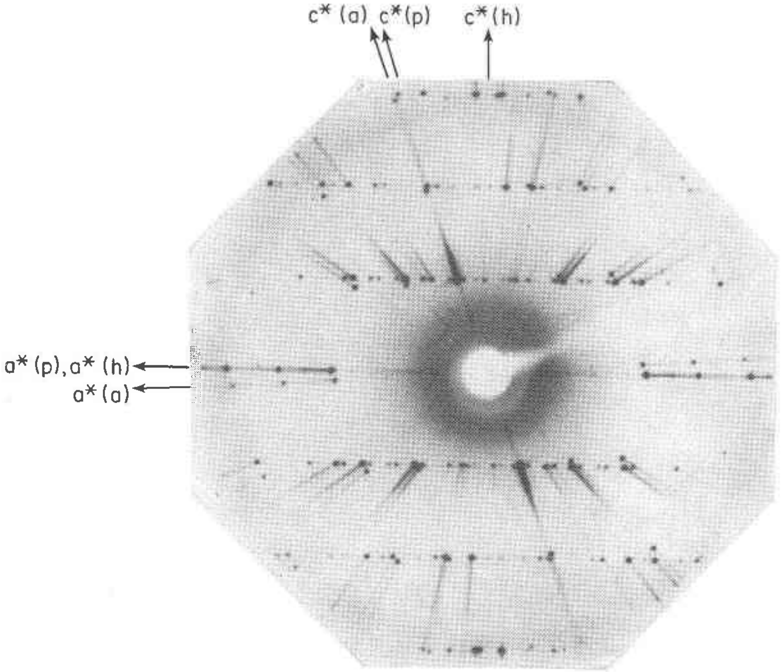

FIG. 3. Precession photograph $(010)_0^*$ of pigeonite from the ferrogabbro 4430, Skaergaard intrusion. MoKα (Zr-filtered), $\mu=30°$, 20 hours. The *a** and *c** axes of pigeonite, augite and hypersthene are indicated. *a**(p), *c**(p): pigeonite; *a**(a), *c**(a): augite; *a**(h): hypersthene.

Table 3. Cell Dimensions and Chemical Composition of Pigeonite, Hypersthene and Exsolved Augite, Coexisting in Pigeonite Crystals from the Ferrogabbro, the Skaergaard Intrusion

Pyroxenes	a(Å)	b(Å)	c(Å)	β(°)	V(Å^3)	Composition
pigeonite (host)	9.69 (1)	8.94 (1)	5.23 (1)	108.8 (1)	428.9	$Mg_{45.5}Fe_{45.9}Ca_{8.6}$ (Brown, 1957)
hypersthene	18.34 (1)	8.94 (1)	5.23 (1)	—	857.5	
augite	9.75 (1)	8.94 (1)	5.26 (1)	105.8 (1)	441.2	

2) respectively, we can conclude that the pigeonites from the two different localities have the same composition. The cell dimensions of the exsolved augite (Table 3) are in good agreement with those of augite (f) (Table 2). Thus the exsolved augite in the pigeonite from the Skaergaard intrusion no longer shares (001) with the pigeonite and exhibits the intrinsic cell dimensions corresponding to its chemical composition. These observations strongly support the assumption and the treatments with the pigeonite from the Moore County meteorite in this investigation.

Strain Energies of the Exsolved Augites

The order of the strain energies obtained for the exsolved augites, 0.02 kcal/mole and 0.04 kcal/mole, is very common for local strains when exsolution or phase transformation takes place in metal and inorganic crystals. These strain energies are too small to produce any mechanical or phase changes in the augites.

The internal stresses in the exsolved augites are derived to be 9 kbar for augite (c) and 14 kbar for augite (a) by applying their strains on the equation (2). Because the crushing strength of rock (say basalt) is about 1.9 kbar in average and ranges over 1.0 to 3.3 kbar (Clark, 1966), the values of stresses mentioned above seem to be apparently large in comparison with the possible crushing strength of common minerals. However, crushing strength is generally much less than intrinsic strength for the usual materials (Epstein, 1948). The internal stresses are, therefore, considered to be reasonable for the exsolved augites. Thus the strain energies and internal stresses can well explain the natural appearance of augite (c) and augite (a) as exsolution products in pigeonite.

Relative Stability of Exsolved Augite in Different Orientations

In the study of the exsolutions of pyroxenes, especially exsolutions of one clinopyroxene (say, Ca-rich) from the other (Ca-poor), Hess (1960) wondered why (001) should have been preferred over (100), (010) or some other plane for the exsolution and suggested some special relationship between the exsolution plane and the ease of migration of ions. The strain energies of exsolved augites in pigeonite can be considered to represent their relative stability, because the structure of augite is almost the same as that of pigeonite (Morimoto and Güven, 1968) and the boundary energies, excluding the strain energies, between the exsolved augites and pigeonite are considered negligible.

The result that E^a is greater than E^c comes essentially from the fact that the direction of the silicon chains, that is, c is the most rigid direction in pyroxene structures as expected from the values of c_{ij} for augite. The difference of E^c and E^a is supported by the facts that (100) lamellae of augite are extremely rare in pigeonite and even if they exist as in the Moore County pigeonite, they are not observed under the microscope though the (001) lamellae are.

Since the strain energies for the exsolved augites on (010) or $\{110\}$ of pigeonite are about ten times greater than E^c and E^a, the exsolution of augite on (010) or $\{110\}$ with such great strain energies are not possible in nature.

EXSOLUTION OF AUGITE (A) IN THE MOORE COUNTY PIGEONITE

When the exsolution of augite takes place in pigeonite under equilibrium conditions, the appearance of augite (a) must exceptional because of its higher strain energy than that of augite (c). In fact, the exsolved augite (a) in pigeonite from the Moore County meteorite is the first and the only example so far described, except that in pigeonites from volcanic rocks (Morimoto, 1966). Therefore, the exsolution of augite (a) must be explained in connection with the formation of the Moore County meteorite.

Hess and Henderson (1948) observed in the meteorites that the crystals of pigeonite and plagioclase tend to lie with their longer axes in one plane but without pronounced linear orientation of the longer axes in that plane. Based on these fabric evidences of the meteorite, they concluded that the pigeonites crystallized very slowly from a large body of magma and accumulated by crystal settling to the floor of magma chamber in an extra-terrestrial body. However if the pigeonites had been cooled slowly as in the Skaergaard intrusion, they should have exsolved only augite (c) with the intrinsic cell dimensions free from lattice distortions.

We can think of two possibilities for the formation of augite (a) in pigeonites from the meteorite. The first possibility is as follows: the pigeonites which had been accumulated by crystal settling exsolved augite, not under hydrostatic pressure, but under uniaxial compression. According to the calculation given above, the strain energy of augite (c) was the same order as that of augite (a) when the pressure on (100) of of pigeonite was stronger by about 4,000 bars than that on (001). The second possibility is as follows: the exsolution of augite took place when the pigeonites were highly over-saturated with augite component because of a sudden change of the cooling rate. Since the difference of the strain energies between augite (c) and augite (a) is small, pigeonites could exsolve both augite (c) and augite (a) when they are highly over-saturated with augite component. Extreme cases are observed in pigeonite from volcanic rocks.

Admittedly these possibilities are speculative and more studies on the phase relations and exsolution phenomena in pyroxenes are needed to understand the speciality of the Moore County meteorite. However, we can conclude that pigeonite from the Moore County meteorite can be considered to represent an intermediate stage in the exsolution process of augite in comparison with pigeonite from the Skaergaard intrusion.

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APPENDIX

The referee has indicated that ϵ_{13} for the exsolved augites should not be omitted at this stage of the calculation for the strains. We have carried out the calculation including ϵ_{13} as follows to check our results in the text.

Substituting the nonzero components into the equation (4), we obtain the following relations, instead of (10), (11), (12) and (13),

$$
\begin{aligned}
c_{13}\overset{c}{\epsilon}_1 + c_{33}\overset{c}{\epsilon}_3 + c_{53}\overset{c}{\epsilon}_5 &= 0\\
c_{15}\overset{c}{\epsilon}_1 + c_{35}\overset{c}{\epsilon}_3 + c_{55}\overset{c}{\epsilon}_5 &= 0\\
c_{11}\overset{a}{\epsilon}_1 + c_{13}\overset{a}{\epsilon}_3 + c_{15}\overset{a}{\epsilon}_5 &= 0\\
c_{15}\overset{a}{\epsilon}_1 + c_{35}\overset{a}{\epsilon}_3 + c_{55}\overset{a}{\epsilon}_5 &= 0.
\end{aligned}
$$

where the relations, $\epsilon_5^a = \epsilon_{13}(a)$ and $\epsilon_5^c = \epsilon_{13}(c)$, are held. From the equation (9), the following relations are obtained,

$$
\begin{aligned}
-\overset{c}{\epsilon}_1 + \overset{a}{\epsilon}_1 &= 0.0087\\
-\overset{c}{\epsilon}_3 + \overset{a}{\epsilon}_3 &= -0.0095\\
-\overset{c}{\epsilon}_5 + \overset{a}{\epsilon}_5 &= -0.0017 + e
\end{aligned}
$$

where e represents a possible error in the measurements.

Using the values of c_{ij}'s for augite in the text, we obtain the following values,

$$\begin{gathered}
\overset{c}{\epsilon}_1 = -0.0059 \\
\overset{c}{\epsilon}_3 = 0.0018 \\
\overset{c}{\epsilon}_5 = 0.0014 \\
\overset{a}{\epsilon}_1 = 0.0028 \\
\overset{a}{\epsilon}_3 = -0.0077 \\
\overset{a}{\epsilon}_5 = 0.0026 \\
e = 0.0029,
\end{gathered}$$

which are in good agreement with the results obtained in the text.