AN X-RAY AND OPTICAL INVESTIGATION OF THE PERISTERITE PLAGIOCLASES

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Abstract

Low-temperature plagioclase feldspars in the peristerite range An₆₊₂ to An₁₇₋₂ unmix on a submicroscopic scale to relieve internal stresses created by substituting Al for Si in the tetrahedral framework. The unmixed components are An₂₆₋₆ and An₂₉₋₃₅, averaging An₃ and An₂₅ respectively. Most peristerites exhibit a bluish schiller as a result of diffusion of light by the submicroscopic domains of different optical polarizabilities. The optical properties of the crystal are weighted averages of those of the domains. Heating at 1000°C causes disordering of Na and Ca between domains, followed by disordering of Si and Al within domains, and final homogenization by disordering of Si and Al between domains. Schiller is retained until the final step of homogenization. Homogenization can not be studied by simple X-ray methods, owing to similarity in unit cell geometry of the high-temperature phases, but can be followed by optic angle measurements. An X-ray technique has been developed to determine the composition of individual unmixed grains, and has been applied to confirm the presence of a break or flexure in the plagioclase refractive index curves near An₁₅, the lower limit of the peristerite region. The upper limit of the peristerite region has not been studied in detail, but deviations from the albite structure sufficient to incorporate the additional Al-content are inferred.

Introduction

The plagioclase feldspars range in composition between the limits albite (NaAlSi₃O₈) and anorthite (CaAl₂Si₂O₈). Once believed to be a continuous solid solution series, the plagioclase series is now known to be structurally discontinuous. Information on the exact mechanism of the substitution of CaAl for NaSi in the series is important in explaining the nature of these discontinuities. This paper deals with a specific discontinuity within the series, located in the peristerite composition range.

As used herein, the term peristerite designates the low-temperature plagioclase feldspars from the albite-oligoclase range that are known to be submicroscopically unmixed into two coexisting plagioclase components and that may or may not exhibit schiller effects. The components have been found to correspond to An₂₆₋₆ and An₂₉₋₃₅ (An₃ and An₂₅ average) and to exist in varying proportions throughout the peristerite range, An₁₅₋₂ to An₁₇₋₂. Unmixing occurs in this range when the 3:1 Si:Al ratio of the low albite composition is exceeded and is believed by this writer to be due to relief of stresses caused by successive replacement of Si by the larger Al, forming minute segregated domains of Si- and Al-rich plagioclase. The Na and Ca cooperate to balance electrostatic valence charges. The ratio of these Ab- and An-rich domains varies

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with total An content of the crystal. Since the two components are submicroscopic in size, the optical properties of the crystal must be weighted averages of those of the components.

Historically, Böggild (1924) was the first to investigate the peristerites. He ascribed their light blue schiller to a reflection phenomenon caused by oriented “labradorizing lamellae” of unstated origin.

Cole, Sörum, and Taylor (1951) in their x-ray study of the plagioclase series mention no deviation from the known low-temperature albite structure, reporting solid solution for the entire range An$_0$ to An$_{30}$. Laves (1951) was the first to note that the schiller of a peristerite was “due to exsolved plagioclase with a geometry slightly different from the geometry of the host.” Laves (1954) expanded his study to show that the unmixing range was An$_8$ to An$_{17}$ and that the two peristerite phases had lattice geometries similar to low-albite and An$_{30}$ respectively. Heated peristerites were homogenized to a high-albite form.

Emmons (1953, revised) in a thorough optical study of analyzed plagioclases gives evidence for a discontinuity in refractive index curves near An$_{18-8}$, the lower limit of the peristerites. An attempt is made herein to discover the nature of this break, which is anticipated from a structural viewpoint. Chayes (1952) shows no such break in his curves, reporting continuous variation of indices with increasing An content. J. R. Smith (1955 b) suggests a number of optical discontinuities, including An$_{11}$ and An$_{22}$, but does not show the An$_{11}$ break in subsequent curves (J. R. Smith, 1957).

J. V. Smith (1956) plotted the change of the reciprocal lattice angle $\gamma^*$ with anorthite content, and Gay and Smith (1955) used the plot to determine that the peristerite phases were actually An$_{32-2}$ and An$_{32-2}$. Most recently, Schneider (1957) investigated the change of lattice geometry of each of the peristerite phases with heat treatment, describing the process of homogenization.

The data on plagioclases used in this study are listed in Table 1.

**Optical Properties of Heated Peristerites**

The optical properties of untwinned cleavage fragments selected from five chemically analyzed plagioclases in the range An$_0$ to An$_{20}$ were determined on the five-axis universal stage as described by Emmons (1943). The $X^*Y^*$ precession photographs (see Fig. 1) showed four of the five specimens to be submicroscopically unmixed. Specimen 7a is structurally homogeneous ($\gamma^* = 88^\circ 34'$, An$_{18-2}$ determined from Fig. 2). Following heating at 1000$^\circ$ C, for various lengths of time (Table 2), the same grains were x-rayed again and were found to have “homogeneous” structures
**Table 1. Specimen Data**

<table>
<thead>
<tr>
<th>No.</th>
<th>Rock Type</th>
<th>Locality</th>
<th>Calculated Wt. %</th>
<th>Source of Analysis*</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>An</td>
<td>Ab</td>
<td>Or</td>
</tr>
<tr>
<td>0</td>
<td>pegmatite</td>
<td>Ramona, Calif.</td>
<td>0.25</td>
<td>98.7</td>
<td>1.1</td>
</tr>
<tr>
<td>1</td>
<td>two-mica granite</td>
<td>Auburn, Maine</td>
<td>5.2</td>
<td>94.0</td>
<td>0.8</td>
</tr>
<tr>
<td>2</td>
<td>biotite granite</td>
<td>Monteagle Tp., Ont.</td>
<td>5.7</td>
<td>90.3</td>
<td>4.0</td>
</tr>
<tr>
<td>3</td>
<td>granite</td>
<td>Villeneuve, Que.</td>
<td>7.6</td>
<td>90.6</td>
<td>1.7</td>
</tr>
<tr>
<td>4</td>
<td></td>
<td>Haddam, Conn.</td>
<td>9.5</td>
<td>90.5</td>
<td>—</td>
</tr>
<tr>
<td>5</td>
<td>two-mica granite</td>
<td>Peekskill, N. Y.</td>
<td>11.4</td>
<td>87.0</td>
<td>1.6</td>
</tr>
<tr>
<td>6</td>
<td></td>
<td>Monteagle Valley, Ont.</td>
<td>13.3</td>
<td>82.8</td>
<td>3.9</td>
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<tr>
<td>7</td>
<td>biotite granite</td>
<td>Parishville, N. Y.</td>
<td>16.1</td>
<td>81.1</td>
<td>2.8</td>
</tr>
<tr>
<td>8</td>
<td>granite</td>
<td>Llano County, Texas</td>
<td>16.6</td>
<td>81.5</td>
<td>1.9</td>
</tr>
</tbody>
</table>

* References: RCE—R. C. Emmons (1953, reprinted 1956); CSR—C. S. Ross in Emmons (1953); CDJ—C. D. Jeffries (1936); VBM—V. B. Meen (1933). Specimens 2, 3, and 6 were contributed by the Royal Ontario Museum of Geology and Mineralogy. Specimens 0, 5, 7, and 8 were contributed by R. C. Emmons.

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**Fig. 1.** A precession photograph of the zero-level X*Y* zone of the peristerite sample 3a. The two distinct X* axes are evidence of the existence of two plagioclases in this specimen. The double Y* and Z* axes are not diagnostic in precession photographs because the reciprocal lattice angles α* and β* vary only 0.2° and 0.1° respectively from An12 to An85. γ* varies by 1.3° in the same range. (J. V. Smith, 1956; Rowbc and Gay, 1958.)

Dotted lines indicate the position of densitometer profiles.
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Fig. 2. Rate of change of $\gamma^*$ with wt. $\%$ An/An+Ab for soda-rich plagioclases. After J. V. Smith (1956).

Table 2. Differences in Refractive Index and Birefringence between High- and Low-temperature Plagioclases

<table>
<thead>
<tr>
<th>No.</th>
<th>Time of Heating</th>
<th>$\gamma^*$ (±5')</th>
<th>$2V_\alpha$</th>
<th>$\Delta$ index of refraction</th>
<th>$\Delta$ birefringence</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>$\gamma$</td>
<td>$\beta$</td>
</tr>
<tr>
<td>0</td>
<td>260 hrs.</td>
<td>88°02'</td>
<td>N.D.</td>
<td>N.D.</td>
<td>N.D.</td>
</tr>
<tr>
<td>2b</td>
<td>112 hrs.</td>
<td>88°15'</td>
<td>58°</td>
<td>-.0020</td>
<td>.0016</td>
</tr>
<tr>
<td>3a</td>
<td>240 hrs.</td>
<td>88°07'</td>
<td>51°</td>
<td>-.0017</td>
<td>.0013</td>
</tr>
<tr>
<td>4a</td>
<td>260 hrs.</td>
<td>88°05'</td>
<td>76°</td>
<td>-.0008</td>
<td>.0015</td>
</tr>
<tr>
<td>6b</td>
<td>425 hrs.</td>
<td>88°01'</td>
<td>42°</td>
<td>-.0021</td>
<td>.0006</td>
</tr>
<tr>
<td>7a</td>
<td>260 hrs.</td>
<td>88°07'</td>
<td>56°</td>
<td>-.0007</td>
<td>.0021</td>
</tr>
</tbody>
</table>
similar to that of high-albite. With one exception (2b) these grains had attained the high-temperature geometry described by J. V. Smith (1956) for the range An₀ to An₆₆ (Fig. 2).

The change in refractive indices recorded for these specimens is in accord with the data of J. R. Smith (1957). It will be noticed that although $\gamma^*$ indicates a high temperature state according to Fig. 2, $2V_\alpha$ for four specimens is not at the minimum to be expected if the disordered distribution of Al-Si is complete for the entire grain. Schneider (1957) has plotted the change of $\gamma^*$ against $2V_\alpha$ with time of heating for a nearly pure albite. This curve, reproduced in Fig. 3, indicates that $2V_\alpha$ is a more

![Fig. 3. A plot of $2V_\alpha$ against $\gamma^*$ for increased time of heating at 1000°C of a nearly pure albite. After Schneider (1957). Small numbers along the curve represent days heated.](image)

reliable indicator of disorder than is $\gamma^*$. The choice of $2V_\alpha$ as an index to the degree of disorder is illustrated by specimen 6b (Table 2). It shows significantly larger decreases in the $\alpha$ and $\gamma$ refractive indices and less increase in $\beta$ after heating than do the other specimens. One would thus expect 6b to be the most completely disordered. This is borne out by the fact that $2V_\alpha$ for this grain is 42°, substantially lower than that for the others. On the other hand, it is impossible to distinguish relative degrees of order from their lattice geometry, for $\gamma^*$ is very nearly equal for all the heated specimens.

In this paper, a detailed study of the change of lattice geometry with time of heating and an investigation of the optical effect known as schiller will lead to a reasonable explanation of the dynamics of the
"homogenizing" and disordering processes operative within the heated peristerite.

**Homogenization** and Disordering of Peristerites

The phenomena of "homogenizing" the separate phases of a peristerite and disordering them were studied in detail by Schneider (1957). Schneider heated an oligoclase (An_{13}) from Amelia Court House, Virginia, and observed the changes of $\gamma^*$ with time of heating at temperatures of 950° C., 1000° C., and 1050° C. Two of his graphs are reproduced in Fig. 4.

The slopes of these curves are steepened by increasing temperature of heating, but a similarity of shape is maintained regardless of temperature. Of particular importance is the observation that the rate of change toward the high-temperature state is greater for the An-rich phase than for the Ab phase. Schneider suggests that the cause of this divergence is a slight degree of disorder that exists in the low-temperature form of the crystal because of its anorthite content. The possibility of slight disorder existing in one of the components is a promising idea in terms of peris-

![Diagram](attachment:image.png)

**Fig. 4.** Plots of the rate of change of the reciprocal lattice angle $\gamma^*$ with heating time (days) at 950° C. and 1000° C. of an Amelia, Virginia, oligoclase. Modified after Schneider (1957). Dotted extensions of Schneider's curves are by this author. A = Ab-rich phase; B = An-rich phase.
terite dynamics. Since the An-rich phase reaches its geometrical high-
temperature state in approximately one-third the time it takes for the
Ab phase to do the same, it is not unreasonable to suppose that the An-
rich phase was already partially disordered, even in the peristerite’s low-
temperature form.

The occurrence of domains of approximately 25% An in all peristerites
(Table 6) is evidence that this structure is singularly stable, even though
Schneider’s findings suggest that it may not be completely ordered with
respect to Al-Si. That structural stability is not necessarily synonymous
with the classical concept of a completely ordered structure was sug-
gested by Ferguson, Traill, and Taylor (1958), who found that a low-
temperature albite they studied had but 72% of the Al in the Si1(O)
tetrahedral site. This albite was calculated to be electrostatically more
stable than the classically ordered albite which theoretically has all the
Al in this tetrahedral site.

If electrostatic balance exerts a more profound influence on plagioclase
structure than has previously been known, it may be found that the low-
temperature An26 unit cell does not have a totally regular Al-Si distribu-
tion taking into account its greater Al content as well as Schneider’s
discovery that the An25 component of an unmixed oligoclase attains a
high-temperature configuration more readily than the albite phase. The
foregoing statements are necessarily speculative, but it is hoped that the
structure of An26, presently being done at Cambridge University, may
clarify this subject.

The dynamics of the “homogenization” of the two components of the
peristerites are imperfectly known. Schneider (1957) does not show the
details of the procedure on his curves of \( \gamma^* \) versus heating time. He re-
ports that homogenization is completed in 170 days at 950\(^{\circ}\) C. and in 35
days at 1000\(^{\circ}\) C. \( \gamma^* = 88^{\circ}05' \) for both homogenized specimens. This infor-
mation is plotted as dotted extensions of his curves in Fig. 4.

Table 3 contains data on peristerites homogenized in the present study
by heating at approximately 1000\(^{\circ}\) C. The reciprocal lattice angle \( \gamma^* \) after
heating varied from \( 87^{\circ}56' \) to \( 88^{\circ}07' \) and averaged \( 88^{\circ}02' \) for nine speci-
mens. Specimens 5a and 6a failed to reach the “homogeneous” high-tem-
perature lattice geometry after an initial \( 7\frac{1}{2} \) day heating at 1000\(^{\circ}\) C.
Table 4 compares the unheated, intermediate, and “homogeneous”
lattice angles with the two phases and gives a rough estimate of the
relative percentage of the Ab phase as judged from the comparative in-
tensities of the white radiation streaks of the X* axes of the two phases.

In order to complete the picture of the disordering and homogenizing

* This figure is subject to a possible error of 25%.
process, the optical effect known as schiller must be considered in detail. Peristerites often exhibit a light to brilliant blue schiller. That schiller is a result of unmixing, at least in the peristerite range for plagioclases and certainly in the perthitic alkali feldspars, has long been assumed. An absence of schiller in unmixed feldspars may indicate that the domains are either too small or too large to cause diffusion of light. A number of investigators have found that under certain conditions some perthites and peristerites retained this schiller after heat treatment (Dittler and Köhler, 1925; Spencer, 1930; Heald, 1950). In this study, specimens 4b and 2a were observed to retain their schiller after 4½ days heating at

<table>
<thead>
<tr>
<th>No.</th>
<th>Wt. % An</th>
<th>Heating time (days)</th>
<th>( \gamma^* ) (±05')</th>
</tr>
</thead>
<tbody>
<tr>
<td>3b</td>
<td>7.6</td>
<td>4½</td>
<td>88°01'</td>
</tr>
<tr>
<td>4b</td>
<td>9.5</td>
<td>4½</td>
<td>88°05'</td>
</tr>
<tr>
<td>6b</td>
<td>13.3</td>
<td>4½</td>
<td>88°01'</td>
</tr>
<tr>
<td>1a</td>
<td>5.2</td>
<td>7½</td>
<td>87°56'</td>
</tr>
<tr>
<td>3a</td>
<td>7.6</td>
<td>10</td>
<td>88°07'</td>
</tr>
<tr>
<td>7b</td>
<td>16.1</td>
<td>10½</td>
<td>88°06'</td>
</tr>
<tr>
<td>4a</td>
<td>9.5</td>
<td>10½</td>
<td>88°05'</td>
</tr>
<tr>
<td>0</td>
<td>0.25</td>
<td>10½</td>
<td>88°02'</td>
</tr>
<tr>
<td>5a</td>
<td>11.4</td>
<td>18</td>
<td>87°56'</td>
</tr>
<tr>
<td>6a</td>
<td>13.3</td>
<td>18</td>
<td>88°01'</td>
</tr>
</tbody>
</table>

(Compare to Schneider's curves in Fig. 4.)
1000° C. X-ray photographs, however, indicated that these same grains had reached a homogeneous high-temperature state.

The explanation for this enigmatic behavior after heating is dependent on analogy with the cause of schiller in moonstones as set forth by Raman, Jayaraman, and Srinivasan (1950). The following is a paraphrase of their significant article:

The blue color seen in moonstones is too rich a blue to be explained as an interference color due to the reflection of light by thin films, but matches the color exhibited in the diffusion of light by particles small in size compared with the wave length of light. The observed facts give no support to the hypothesis of a lamellar structure as the cause of schiller [contrary to Böggild, 1924]. Diffusion is substantiated by the color and the fact that schiller “reflections” are visible through a wide angle (14 to 18°).

Fluctuations of the refractive index consequent on the local fluctuations in chemical composition (and resultant structural adjustments) would give rise to a strong diffusion of light, its intensity increasing in proportion to the number of clusters, multiplied by the square of the volume of each cluster [of Na as segregated from K in the continuous tetrahedral framework of the feldspar]. This is possible provided that these clusters or “crystallites” are of sufficiently small dimensions and are not co-planar. These crystallites must be mutually oriented.

Since the diffusion of light in its passage through the crystal is a consequence of optical heterogeneity, its intensity depends on the magnitude of the differences in optical polarizabilities of the separate domains. Because birefringences are different for different directions in the crystal, the schiller effect varies.

With the exception of the degree of symmetry change between domains, the peristerites can be shown to fit the moonstone picture ideally. Table 5 shows that the differences in refractive indices for Or-Ab (moonstones) and for Ab-An* (peristerites) are of the same order of magnitude. This in itself appears to be sufficient explanation for the existence of schiller in low-temperature peristerites, which are known to be unmixed on a submicroscopic scale and segregated into domains of Ab- and An-rich plagioclases.

The retention of schiller in heated and supposedly homogenized peristerites can be explained in the same manner, for the high-temperature Ab and An* phases have differences in refractive indices comparable to those of the low-temperature phases (Table 5). Homogenization has been assumed from the X*Y* x-ray precession photographs where only one X* axis—not two as in unmixed grains (Fig. 1)—is seen making an angle γ* = 88°02' ± 06' with the single Y* axis. Because schiller persists in these “homogenized” grains, it is now suggested, following Laves (1952) and Hewlett (1959), that the domains actually still exist in the heated specimen, causing optical heterogeneity that in turn gives rise to diffusion of light or “schiller.”

Inspection of the X*Y* zone of heated but not yet “homogenized” grains 5a and 6a (Table 4) shows that the reciprocal lattice parameter a*
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Table 5. Differences between Refractive Indices of Domains in Moonstones and in Low- and High-temperature Peristerites

<table>
<thead>
<tr>
<th>Domain composition</th>
<th>Refractive index ($N_D$)</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\gamma$</td>
<td>$\beta$</td>
</tr>
<tr>
<td>Low-temp. Or Ab</td>
<td>1.5245</td>
<td>1.5228</td>
</tr>
<tr>
<td>Difference:</td>
<td>.0147</td>
<td>.0100</td>
</tr>
<tr>
<td>Low-temp. Ab $\text{An}_{25}$</td>
<td>1.5379</td>
<td>1.5314</td>
</tr>
<tr>
<td>Difference:</td>
<td>.0107</td>
<td>.0133</td>
</tr>
<tr>
<td>High-temp. Ab $\text{An}_{25}$</td>
<td>1.5354</td>
<td>1.5342</td>
</tr>
<tr>
<td>Difference:</td>
<td>.0124</td>
<td>.0114</td>
</tr>
</tbody>
</table>

has become equal or very nearly so for the two partially disordered components. This may indicate that Na and Ca are randomly distributed throughout the crystal at this stage of heating, although two phases differing in Si-Al ratio still exist (Laves, 1952, p. 567). With further heating, the two $X^*$ axes come together and coalesce as described by Schneider (1957), apparently indicating homogenization to a single phase crystal. It is proposed, however, that there are remnant domains that retain a high degree of their original Al–Si content and are disordered with respect to Al–Si, within themselves. Because they contain differing amounts of Al–Si, they still have different refractive indices and, therefore, still cause schiller. Heating has caused disordering of Si–Al within, but not between, domains at this stage.

It is important to note that both the Ab and the $\text{An}_{25}$ domains, now in the high-temperature state, have nearly identical lattice parameters (J. V. Smith, 1956). This geometrical similarity means that these phases are not distinguishable by simple x-ray techniques and thus give the erroneous impression of homogeneity. Optical heterogeneity persists, for the differences in refractive indices for high-temperature forms of Ab and $\text{An}_{25}$ are of the same order of magnitude as those causing moonstone schiller.

True homogenization demands a random distribution of Al–Si between as well as within domains, thereby eliminating the domains. Specimens 2a and 4b and six schillered grains from sample 1 lost their schiller
only after 35 days heating at 1000° C. Two other grains from sample 1 required additional heating at 1050° C. to destroy the remnant domains. Duration of the heat treatment necessary to disorder completely and thus truly homogenize peristerites varies widely from sample to sample.

In the course of investigation of these chemically analyzed peristerites, the limits of unmixing (An₅ and An₁₇) suggested by Laves (1954) were confirmed. The lower boundary of the two-phase peristerites was established by chips from sample 1. These chips were selected from the same region of a 75 gm. schillerized single crystal (An₅.₂Or₀.₈). Of the eight chips chosen, four were found to be structurally homogeneous and four were two-phase peristerites. Pertinent structural data are listed in Table 6. If the chemical analysis of this specimen is accurate, the boundary below which structural homogeneity exists can be fixed at An₆±₂.

Six grains from sample 7 (An₄₆.₁Or₂.₈) were found to have homogeneous structures in their natural state. The composition of these grains determined by x-ray techniques described below is An₁₅. Laves (1954), however, claims to have observed unmixing in grains from this same sample (his #8). A single crystal from specimen 8 (An₁₆.₆Or₀.₉) proved to be unmixed. Lattice measurements indicate that the two phases correspond to An₅ and An₂₆. Since at least some grains from sample 7 are single phase and some from sample 8 (Laves' #9) are peristerites, the upper limit of structural inhomogeneity can be established at An₁₇±₂.

Specimen 8 is interesting also in that it could be determined that the An₃ phase is either twinned by the Albite law or has two orientations within the crystal characteristic of this twin law, whereas the An₂₆ phase is untwinned.

Refractive Index Curves

Although there is agreement on the limits of the peristerite range, considerable differences exist in the published optic curves for this composition region. Consequently, a detailed study of this region demands x-ray, optical, and compositional data on individual grains.

For this purpose an x-ray method was devised to determine An content for individual unmixed grains, thereby permitting closer correlation of optics and composition than is possible with bulk chemical analysis. It is a moderately successful method, applicable only to low-temperature soda-rich plagioclases.

Case I. Homogeneous low-temperature plagioclase

The angle γ* is measured from the X*Y* x-ray precession photograph and checked against J. V. Smith's (1956) plot of γ* versus An content (Fig. 2). The accuracy of angular measurement is usually better than 0.5'
Table 6. Optical and X-ray Data for Selected Peristerites

Only directly determined indices of refraction and those whose rotated and calculated (from 2V) values are equivalent are recorded.

<table>
<thead>
<tr>
<th>No.</th>
<th>Chem. analysis (wt. %)</th>
<th>Refractive indices (ND)</th>
<th>&quot;Strong&quot; phase</th>
<th>&quot;Weak&quot; phase</th>
<th>% An determined from refractive indices</th>
<th>X-ray</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>An</td>
<td>Or</td>
<td>n.d.</td>
<td>1.5368*</td>
<td>1.5329</td>
<td>γn</td>
</tr>
<tr>
<td>0</td>
<td>0.25</td>
<td>1.1</td>
<td></td>
<td>1.5369†</td>
<td></td>
<td>90°29'</td>
</tr>
<tr>
<td>1a</td>
<td>5.2</td>
<td>0.8</td>
<td>1.5398</td>
<td>1.5300</td>
<td>90°24'</td>
<td>5.0</td>
</tr>
<tr>
<td>1b</td>
<td>5.5</td>
<td>0.5</td>
<td>1.5400</td>
<td>1.5284</td>
<td>90°22'</td>
<td>2.5</td>
</tr>
<tr>
<td>1c</td>
<td>5.1</td>
<td>0.3</td>
<td>1.5395</td>
<td>1.5336</td>
<td>90°28'</td>
<td>0.5</td>
</tr>
<tr>
<td>1d</td>
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<td>0.4</td>
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of arc, and composition may be quoted to ±2% An for the range An₀ to An₂₃. This does not take into account errors in the graph itself. From An₂₃ to An₄₀, precision is not better than ±3% An.

Case II. Two-phase (unmixed) low-temperature peristerites

The angles γ* are measured from the single Y* axis to both X* axes. It is important that the strong and the weak phases be designated. Using unfiltered χ-radiation, care is taken to obtain maximum contrast of X* streaks against the film. A fine grained film and the largest permissible precession angle are suggested. Finger prints or scratches along the profile to be measured for intensity are highly restricting and may render the film useless.

A densitometer is used to obtain an intensity profile across the X* white-radiation streaks at a position of greatest resolution. The galvanometer deflection, directly proportional to the X* axis intensity, is plotted against film movement as in Fig. 5. A convenient method of determining peristerite single crystal composition is given by the formula:

\[
\frac{\text{Area}_A}{\text{Area}_{(A+B)}}\cdot(\text{wt. }\% \text{ An}_A) + \frac{\text{Area}_B}{\text{Area}_{(A+B)}}\cdot(\text{wt. }\% \text{ An}_B)
\]

with reference to Fig. 5. Anorthite contents determined by this method are shown in Table 6 and Fig. 7.

The method for unmixed grains may well be questioned regarding its cumulative error. Contrasting it with An-contents determined by optics.
and by chemical analysis, the deviations amount at most to \( \pm 2.5\% \) An. This is not a true test of accuracy, however, since deviations from the bulk chemical analysis may only reflect chemical variations within the sample, which may take any value. Existing optical curves are also based on bulk chemical analyses and depend for their validity on an adequate sampling and averaging technique. Considerable differences exist between published curves for this composition region. These factors affect accuracy of the x-ray method: (1) background darkening from scattered radiation, (2) degree of resolution of white radiation streaks, (3) error of measurement of \( \gamma^* \), and (4) accuracy of Smith's (1956) curves, which also require an adequate sampling and averaging technique for their validity. There is an occasional chance that the \(+X^*\) axis intensity profile does not show exactly equivalent area ratios as the profile over the \(-X^*\) axis. Averaging will minimize an otherwise small error.

At this stage of development, this method cannot claim the desired \( \pm 2\% \) accuracy. It is suggested that this system be used with caution and due regard to the variables, and it is hoped that refinements that will both expedite and amend the procedure can be developed. The method fails for a peristerite in an intermediate thermal state, for \( \gamma^* \) changes with the degree of order-disorder and thus cannot be used as an index to composition. Since the high-temperature modifications of all soda-rich plagio-

Fig. 5. Plot of the traverse in mm. of resolved \( X^* \) axes of peristerite 5f against the intensity of their white-radiation streaks in units of galvanometer scale deflection. The method of determining peristerite single crystal composition is given by the formula:

\[
\frac{\text{Area}_A}{\text{Area}_{(A+B)}} \cdot (\text{wt. } \% \text{ A}_A) + \frac{\text{Area}_B}{\text{Area}_{(A+B)}} \cdot (\text{wt. } \% \text{ A}_B)
\]
Plagioclases have very similar geometry, they are indistinguishable one from the other by ordinary x-ray techniques.

Using these methods, compositions may be assigned to individual grains in the low-temperature state. The value of these devices becomes obvious when the refractive index curves plotted against analyzed bulk compositions are contrasted with those plotted against An content determined by x-ray techniques for individual grains (Figs. 6, 7).

Refractive indices of thirty-three grains from eight analyzed low-temperature plagioclases are recorded in Table 6 along with pertinent x-ray data. The average indices of these samples are plotted according to bulk chemical analyses in Fig. 6. Figure 6 also contrasts the plagioclase index curves of Emmons (1953, revised) and Chayes (1952). The experimental points fall between the two curves, although slightly closer to Emmons' curves and confirming a break or flexure in the 5% An region. This break is to be expected from a structural viewpoint at the lower boundary of the unmixed peristerite range, and further evidence for it appears in Fig. 7 where individual indices are plotted against wt. % An/An+Ab of individual grains determined by the x-ray methods described above. Of particular interest is sample 1. It contains both unmixed, two-phase grains and structurally homogeneous grains. Although chemical analysis
PERISTERITE PLAGIOCLASES indicates An₆.₃Or₀.₈ to be the bulk composition, x-ray methods show a range from An₀.₅ to An₆.₇ for individual fragments. The homogeneous grains range from An₀.₅ to An₆; unmixed grains range from An₃.₅ to An₆.₇.

It was expected that all the unmixed grains would have higher refractive indices than the structurally homogeneous grains of the same composition, for unmixing can be considered an extreme case of ordering, representing a closer packing of ions, increased polarization, and higher refractive index. But this is not the case for sample 1. For example, grain 1b is unmixed and its two components, approximately An₆ and An₆₆, are present in the ratio 94.₄/5.₆ (x-ray method) and give a total composition of An₆.₇. This grain has the same indices as grain 1h, which is homogeneous and contains approximately 6% An according to γ* measurements. Further contrasted with 1h is the homogeneous grain 1d whose reciprocal lattice angle γ* indicates a composition of only An₀.₅. Yet the indices of 1b, 1h and 1d are nearly identical within the limits of error of measurement (±₀.₀₀₀₄).

Because γ* varies so widely for these homogeneous grains and because both structurally homogeneous and unmixed grains exist in the same crystal, a significant variation in An content from grain to grain is unquestioned. The plot of the refractive indices of the grains from this crystal against composition as determined by the x-ray method (Fig. 7)

![Refractive indices plot](image-url)

**Fig. 7.** Refractive indices of twenty-three grains from eight low-temperature soda-rich plagioclases plotted against wt. % An/An+Ab determined by X-ray methods. All specimens of less than 7% An are from sample 1. Lines of best fit are indicated.
demonstrates that some sort of break or flexure in index curves is present in the An₅ to An₁₀ region. The break here falls at An₇ whereas in Fig. 6, using average indices and bulk chemical analyses, the break falls at An₅. Emmons (1953, revised) indicated a break between An₅ and An₇ in his determinative chart of plagioclase optics. Neither Chayes (1952) nor J. R. Smith (1957) shows a change of slope here.

Alternate explanations of these unusual properties include nonequilibrium conditions and factors other than composition that affect the γ* parameter. At present there is insufficient evidence to permit intelligent speculation on either of these possibilities.

The anomalous optical and structural situations in the 5% An region attest the necessity of a re-evaluation of this composition range. More analyzed specimens and an improved method of determining composition of individual grains are indispensable to a thorough understanding of the peristerite plagioclases. The possibility of another break of flexure in the optic curves at the other end of the peristerite range as suggested by J. R. Smith (1957) at An₃₁, was not examined in this study.

**DISCUSSION**

The cause of peristerite unmixing as a soda-rich plagioclase cools during crystallization is not certain, although in the case of the somewhat similar moonstones or crypto-perthites it is assumed to be due to stresses caused by the 35% discrepancy in ionic radii of K and Na. It is proposed here that the original homogeneous crystal of peristerite composition is caused to unmix during cooling by the large size difference between Al and Si in the feldspar tetrahedral framework. This size difference is 38% based on radii of 0.29 Å and 0.47 Å for Si and Al respectively. The radii are determined from the data on feldspar bond lengths as listed below. The O–O distance was obtained from the data of Bailey and Taylor by plotting their O–O bond lengths versus Al content for the individual tetrahedra and extrapolating to zero Al content. It is assumed that the oxygens are in contact at this composition.

\[\begin{align*}
\text{Si-O} & : 1.60 \text{ Å (J. V. Smith, 1954)} \\
\text{Al-O} & : 1.78 \text{ Å (J. V. Smith, 1954)} \\
\text{O-O in SiO₄ tetrahedron} & : 2.61 \text{ Å (Bailey & Taylor, 1955)}
\end{align*}\]

This 38% size difference contrasts with an 8% variance between Na and Ca in octahedral coordination \(r_{Na} = 0.98 \text{Å}; r_{Ca} = 1.06 \text{Å}\). The driving force for the unmixing can thus be reasonably associated with the Al–Si, the Ca–Na cooperating to balance the electrostatic valence charges.

The albite structure can tolerate only minor deviations from the composition Na₂Al₅Si₃O₁₀, for with increasing substitution of the larger Al for Si, strains are established in the crystal lattice that can be relieved only
by a different distribution of the misfit atoms—changing from a random
distribution of the Al in excess of the geometrically stable 1:3 Al:Si ratio
within the tetrahedral sites of the homogeneous crystal to a segregation
into Al- and Si-rich regions. It is presumed that the framework is con-
tinuous throughout the two-phase peristerite, but distorts elastically
within the individual domains to assume the geometry of the local com-
position, namely An$_3$ or An$_{58}$. This view infers that the structure of An$_{58}$
is enough different from that of pure albite to tolerate the additional Al
content, perhaps by an ordered geometry different from that of albite. It
is presumed that the unmixed components remain submicroscopic in
size, in contrast to perthites, because of the difficulty of diffusion of Al
and Si over any appreciable distance as a consequence of the strength of
their bonds (Goldsmith, 1952).

**Acknowledgments**

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