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The Peristerite Problem

By *K. Viswanathan* and *E. Eberhard* (Zürich)*)

With 4 figures and 3 tables in the text

Zusammenfassung

Pulver- und Einkristallaufnahmen an 17 Peristeriten ergaben: 1. Peristerite mit einer Pauschalzusammensetzung zwischen 8 und 16% An weisen im allgemeinen drei „Phasen“ auf. 2. Die An-reichste Phase zeigt im Pulverdiagramm stets verschwommene Linien, welche auf eine Variation im Chemismus dieser „Phase“ schliessen lassen. 3. Die Temperatur für die Peristeritentmischung liegt unterhalb von 500° C. Die Kinetik des Entmischungsvorganges wird durch die Annahme einer Schar von Entmischungskurven erklärt.

Abstract

Powder- and precession photographs made from seventeen samples of peristerites lead to the following observations: 1. Specimens with bulk compositions lying between 8% An and 16% An show in general three phases. 2. The reflections of the phase richest in anorthite content are always diffuse indicating a range of compositions. 3. The peristerite unmixing takes place below 500° C. The kinetics of the unmixing process is explained by assuming a series of exsolution curves.

LAVES (1951, 1954) first suggested that peristerites consist of two coexisting plagioclase phases, the compositions of which approach An_0 and An_{30} . He further showed that the peristerite exsolution can be observed in plagiocalses having a bulk composition between An_5 and An_{17} . GAY and SMITH (1955) suggested the compositions $An_{3\pm 2}$ and $An_{23\pm 2}$ for the respective end members. BROWN (1960) established that even albites having the composition 2% An show unmixing and he gave the compositions of the end members as 0% An and 25—28% An respectively. RIBBE (1960), RIBBE and VAN VOTT (1962) and RIBBE (1962) investigated the problem further and suggested an asymmetrical exsolution curve for peristerites. RIBBE (1962) noted a relationship between the

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bulk composition of the peristerites and the composition of the An-rich phase of the exsolved material. NISSEN and BOLLMAN (1966) have suggested on the basis of an electron-microscopic study of peristerites that the composition of the An-rich phase is approximately 18% An.

While working on the plagioclases of Ticino, Switzerland, the authors came across a few specimens whose compositions were found to lie in the peristerite range (0%—17% An). X-ray powder photographs were made of all these specimens and it was observed that four of them showed "Peristerite-unmixing", and the others were homogeneous. It was noted that homogeneous crystals (with a slightly different composition) also occurred in the same hand specimens in which peristerites were found. These new observations made it worthwhile to study the peristerites in detail. Thirteen other specimens from different parts of the world were also included for this investigation.

A. NIGGLI (1967) concluded on the basis of theoretical considerations that the positions possible for Al atoms in an acid plagioclase of some oligoclase composition must be different from those of a pure albite. This means that it is not possible to produce a plagioclase of composition 25% An (at which composition, according to Niggli, an ordered structure is possible) starting from an albite structure by simple replacement of silicon atoms, and hence a change in the structure must take place¹).

GOLDSMITH and LAVES (1961) have stated, in connection with the exsolution of alkali feldspars, that the different Al/Si-distributions, which characterise the different degrees of order, would give rise to different exsolution curves. MUELLER (1968) found that disordered alkali feldspars gave homogeneous crystals above 700°C whereas ordered alkali feldspars could be homogenised only at about 900°C. Thus there is evidence indicating that the exsolution temperature and the type of exsolution curve are considerably influenced by the nature of distribution of the Si and Al atoms in the structure of the feldspar undergoing exsolution.

In the case of plagioclases, different types of Al/Si distributions are caused not only by order/disorder phenomena, but also by changes in anorthite content, which give rise to different Si/Al-ratios. It must, therefore, be realised that the peristerite exsolution is influenced by both these factors. As it is shown later that the exsolution takes place at low temperatures (lower than 500°C) and hence the distribution of Si and Al atoms is nearer to the ordered than to the disordered state, differences in the chemical compositions may be expected to have a greater influence on the exsolution process than order/disorder phenomena.

¹) Ribbe (personal communication to Prof. F. Laves) has determined the structure of a plagioclase of composition An₃₀ and has found that three tetrahedral sites have 0.20 Al, 0.21 Al and 0.22 Al, while the fourth has 0.66 Al.

As feldspars can also form metastably at low temperatures in a disordered state and as such specimens during the process of ordering will have to pass through a series of unstable Al/Si-distributions, the following cases of exsolution must be distinguished:

a) The crystal unmixes with a *stable* Al/Si-distribution.

This means that it must have formed at a temperature higher than that at which the exsolution starts and assumed a stable Al/Si-distribution consistent with that particular temperature. It must have cooled slowly and exsolved.

b) The crystal unmixes with *unstable* Al/Si-distribution.

Here again there are two possibilities.

1. The crystal must have formed at a temperature lower than the probable temperature of exsolution.
2. It must have cooled from a higher temperature very rapidly; as a result it might have an unstable Al/Si-distribution, when it starts unmixing.

In both these cases the Al/Si-distributions could not have been identical with those of the case (a) and the exsolution could not have started at the equilibrium temperature corresponding to that composition.

METHODS OF STUDY

Powder photographs were made of all the specimens from Ticino with $\text{Cu-K}\alpha_1$ and a Guinier-type camera (AEG, Germany). Moreover, $\text{Fe-K}\alpha_1$ radiation was used to take powder photographs of the four peristerites. The differences in the 4θ -values of the line pair $131/1\bar{3}1$ were measured (Table 1). In the case of the unmixed specimens only those lines of the individual phases are resolved (in powder photographs) which show considerable displacement with slight variations in the chemical compositions. Of the pair $131/1\bar{3}1$, the line best suited to show a resolution is $1\bar{3}1$; in contrast the resolution shown by 131 is usually poor. The $24\bar{1}$ reflection of the An-rich phase usually overlaps with the line $31\bar{2}$ of one of the other phases. The An-rich phase, especially, showed in most cases very diffuse reflections; as a result only average measurements could be made.

Precession photographs of the four peristerites from Ticino showed very diffuse reflections with tails indicating a mosaic structure.

The chemical composition (Table 1) was determined with the aid of an electron microprobe (ARL) using small crystal fragments.

A slightly different procedure was adopted to study the thirteen other peristerites included for this investigation. (These specimens originate from the

Table 1. *Chemical data and $(4\theta_{131}-4\theta_{\bar{1}\bar{3}\bar{1}})$ -values ($Fe K \alpha_1$) of homogeneous plagioclases from Ticino (Switzerland)*

Sp. No.	Locality	Composition			$(4\theta_{131}-4\theta_{\bar{1}\bar{3}\bar{1}})$	$(4\theta_{131}-4\theta_{\bar{1}\bar{3}\bar{1}})$ (Corr. *)
		An	Or	Si/Al-ratio		
1729	Preonzo	0.4	0.2	2.97	2.34	2.34
1738	Claro-Monastero	0.5	0.2	2.97	2.27	2.27
1731	Gordola	6.6	0.7	2.75	2.47	2.49
1746	Mogheno-Aurigeno	9.2	0.2	2.66	2.75	2.75
1725	Lodrino-Prosito	9.9	0.4	2.64	2.70	2.71
1737	Claro-Monastero	10.1	2.0	2.64	2.57	2.62
1754	Corcapolo-Intragna	10.9	1.4	2.60	2.68	2.72
1728	Preonzo	11.2	0.8	2.60	2.64	2.66
1748	Ponte Brolla	11.7	0.8	2.59	2.79	2.81
1763a**	Palagnedra	11.9	2.1	2.58	2.70	2.75
1764a**	Slogna-Corcapolo	12.0	1.1	2.58	2.79	2.82
1723	Bonirolo-Mairano	12.7	0.6	2.55	2.77	2.78
1735	V. Verzasca (Ör)	12.8	0.5	2.55	2.85	2.86
1730	Lower Sementina Valley	14.6	0.9	2.49	2.95	2.97
1752	Verdasio	15.2	1.3	2.47	2.91	2.94
1753b**	Verdasio	15.4	2.6	2.47	2.83	2.89
1750	Camedo	16.5	0.8	2.43	2.88	2.90
1740	Cevio	17.9	0.5	2.39	3.21	3.22
1736	V. Verzasca (Ör)	18.9	0.8	2.34	3.13	3.15
1762c**	Palagnedra	19.4	2.8	2.34	3.05	3.12

* Corrected for Or-content.

** Homogeneous oligoclase crystals which occur associated with the peristerites in the same hand specimens.

collection of plagioclase samples listed by CORLETT and EBERHARD (1967) and hence the same numbers given in their table are assigned to these thirteen specimens.) A piece weighing about 30 mg was taken from each specimen. It was broken into three small pieces (each weighing about 10 mg) and three powder photographs were made with the Guinier-camera using $Cu-K \alpha_1$ radiation. In most cases it was found that the three photographs showed differences in the relative proportions of the different phases as judged by the relative intensities of the reflections. This leads to the conclusion that the peristerites are in general inhomogeneous. Hence in order to obtain the true proportions of the different phases a fairly large amount (2—15 gms) of each specimen was powdered and thoroughly mixed. A small portion (10 mg) of it was used for powder photographs with $Fe-K \alpha_1$ radiation. The lines $1\bar{3}\bar{1}$ and $24\bar{1}$ of the individual phases were identified. The 4θ -values of these reflections were measured. In a few photographs the lines of the albite-rich phase were so intense and sharp that even the lattice constants could be calculated using about 40 reflections; the refinement was carried out on the CDC 1604-A computer, using a least-squares refinement program (ZH-LCLS).

A portion of the remaining powder was fused and glasses were prepared for each specimen. These were reground, melted and cooled twice in order to obtain better homogeneity. Then their chemical compositions were analysed with reference to Ca and K using an electron microprobe. It must be noted that

Table 2. *Chemical Compositions and X-ray Data of Peristerites*

Sp. Nr.	% An	% Or	$4\theta_{131} \text{ Fe K}\alpha_1$			$4\theta_{241} \text{ Fe K}\alpha_1$			γ^*		
			Ab-rich phase	Third phase	An-rich phase	Ab-rich phase	Third phase	An-rich phase	Ab-rich phase	Third phase	An-rich phase
137	4.0	1.2	76.18°	—	...	88.91°	—	...	90°19'	—	89°04'
202	4.6	0.7	76.19°	—	75.47°	88.99°	—	...	90°21'	—	89°09'
99	5.0	0.1	76.22°	—	75.49°	88.91°	—	...	90°17'	—	89°13'
203	8.0	1.5	...	75.85°	75.31°	88.83°	89.27°	89.82°	89°56'	—	89°10'
172	8.2	0.3	76.22°	—	75.56°	88.94°	—	89.72°	90°18'	—	89°03'
118	8.5	0.4	76.25°	75.90°	75.51°	88.95°	89.26°	89.64°	90°13'	...	89°12'
187	9.0	0.6	76.22°	75.94°	...	88.96°	89.23°
129	9.6	0.5	76.34°	75.94°	75.40°	88.97°	89.22°
14	12.0	1.4	76.20°	75.88°	75.16°	88.70°	89.29°	...	90°20'	89°45'	89°12'
102	12.0	0.6	76.45°	75.95°	75.50°	88.87°	89.28°	89.70°	90°20'	—	89°14'
135	12.6	0.4	76.25°	—	75.35°	88.95°	—	89.63°	...	—	...
165	12.6	1.1	76.15°	75.80°	75.50°	89.06°	89.36°	89.77°
127	12.7	0.8	76.19°	75.72°	...	88.99°	89.40°
1764b	11.1	1.1	76.22°	75.90°	75.24°	89.00°	89.22°
1762a	12.4	1.9	...	75.89°	75.49°	...	89.35°	89.67°
1762b	12.4	1.8	...	75.84°	75.47°	...	89.38°	89.67°
1763b	13.2	2.0	76.15°	75.84°	75.63°	89.18°	89.31°
1753	15.3	2.4	...	75.70°	75.53°	...	89.41°
			(010)/(101)			(010)/(424)			(010)/(424)		
137	4.0	1.2	86°02'	—	87°08'	72°07'	—	73°26'	79°40'	—	78°28'
202	4.6	0.7	85°58'	—	87°13'	72°08'	—	73°15'	79°39'	—	78°39'
99	5.0	0.1	85°56'	—	86°56'	72°06'	—	73°16'	79°30'	—	78°40'
203	8.0	1.5	...	—	...	72°14'	—	73°01'	79°30'	—	78°45'
172	8.2	0.3	85°50'	—	87°04'	71°58'	—	73°09'	79°45'	—	78°42'
118	8.5	0.4	85°58'	...	87°00'	72°09'	...	73°19'	79°36'	...	78°34'
187	9.0	0.6
129	9.6	0.5
14	12.0	1.4	...	86°19'	...	72°03'	72°29'	73°04'	79°41'	79°12'	78°51'
102	12.0	0.6	...	—	...	72°02'	—	73°03'	79°45'	—	78°50'
135	12.6	0.4	...	—	—	73°10'	...	—	78°45'
165	12.6	1.1	71°55'	...	72°56'	79°30'	...	78°50'

... Phase present but not measurable — Phase not observed in the photograph

Table 3. *Anorthite Contents of the An-rich Phases derived from the different Parameters given in Table 2*

Sp. No.	$4\theta(131)$	$4\theta(241)$	γ^*	010/101	010/424	010/424
137	25.0	26.8	31.8	31.6
202	23.0	23.0	28.4	27.0	26.6
99	22.4	22.3	22.3	26.8	26.2
203	27.6	31.2	24.0	22.2	23.8
172	20.6	28.8	23.0	25.0	24.6	25.4
118	22.0	26.2	22.4	22.8	28.6	29.0
187
129	25.0
14	32.2	22.4	23.0	21.8
102	22.2	28.2	22.1	22.8	22.2
135	21.4	26.0	25.0	24.0
165	22.2	30.6	20.4	22.2
127

the glasses contain all the antiperthitic potash feldspars, which might have also been homogenised. To correct for this and to determine the exact An-content the K-content in the matrix of the crystals of each specimen was determined.

Sections parallel to (001) were made and examined under the microscope. A few of them (specimens Nos. 127 and 135 in Table 2) showed somewhat regularly spaced, cloudy (more or less altered) patches. Small crystals were chipped off from each section for taking precession photographs. All the crystals were mounted with the b-axis as the goniometer axis and pictures were made with [101]—and *c*-axis as precession axis using Cu-K α radiation. The angles γ^* , $010/\bar{1}01$, $010/\bar{4}24$, $0\bar{1}0/4\bar{2}4$ were measured. Precession photographs were also made from crystals separated from the “cloudy patches”. The results are given in Table 2.

RESULTS

1. The differences in the 4θ -values of the line pair $131/\bar{1}\bar{3}1$ of the homogeneous material from Ticino were plotted in the diagram of EBERHARD (1967) with a view to determine their structural state (Fig. 1). It is observed that all the plots lie below the curve representing 500°C even when they are corrected for their Or-content. It may therefore be assumed that the peristerite unmixing

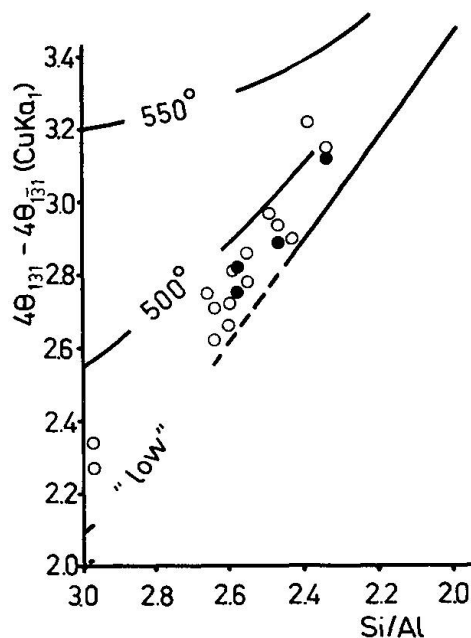


Fig. 1. Diagram to determine the structural state of plagioclases from Ticino, Switzerland. The plotted values of $4\theta_{131} - 4\theta_{\bar{1}\bar{3}1}$ are corrected for their Or-content. The solid circles represent the specimens which occur associated with the peristerites in the same hand specimens.

takes place below 500°C. Moreover, Fig. 1 shows many specimens with bulk compositions lying in the peristerite range (Si/Al ratios lying between 2.97 and 2.45). But it is noteworthy that these specimens, though homogeneous, possess $4\theta_{131-1\bar{3}1}$ values which fit the "plagioclase (low)" curve interpolated between An_1 and An_{17} . This fact seems to suggest that peristerite unmixing does not take place readily; perhaps some additional factors act as catalysts.

2. Peristerites give two kinds of powder photographs, one of them showing only two phases (an albite-rich and an anorthite-rich) and the other indicating the presence of three phases. Specimens having a bulk composition of 0—8% An are observed to show the former, while those richer in anorthite (8—15% An) may give the latter type. The three phases indicated in the second type of photographs are found to be an albite-rich phase, an anorthite-rich phase and a "third" phase which (see later) is found to have a composition of about 16% An. Furthermore those specimens which showed the presence of "cloudy" patches in thin sections showed lines which can be attributed to the presence of zoisite. The zoisites must have been produced by partial alteration of the An-rich phase. In such cases one gets an exaggerated amount of the Ab-rich phase. However in other cases in which there was no evidence for such an alteration, the proportion of the albite-rich phase to anorthite-rich phase, as judged by the relative intensities of the reflections, is found to depend upon the bulk composition of the peristerites. Till about 8% An the albite-rich phase was always predominant.

3. In many cases the agreement between the data collected from the precession photographs and the powder photographs is not very satisfactory. For example the precession photographs of the peristerites of bulk compositions greater than 8% An do not always indicate the presence of three phases. But it must also be realised that the amount of material used for a precession photograph is approximately $1/500$ times that used for a powder photograph and hence the probability of finding all the three phases within such a small portion of the crystal is little, especially when one considers the inhomogeneity of the peristerites.

The precession photographs taken from the "cloudy" patches referred to above show very diffuse reflections and indications of a mosaic structure.

4. The different data collected from the powder- and precession-photographs are plotted in several diagrams in Fig. 2. The working curves were calculated using the lattice constants of plagioclases (low) given by BAMBAUER et al. (1967). It is clearly seen that the same specimen shows different amounts of deviations in the different diagrams. Mostly the deviations fall outside the limits of experimental errors. These discrepancies must, therefore, be attributed to distortions of the lattices of the individual phases. Hence it is impossible to determine the exact An-content of the unmixed phases by X-ray methods.

5. Determinations of the lattice constants of two Ab-rich phases show

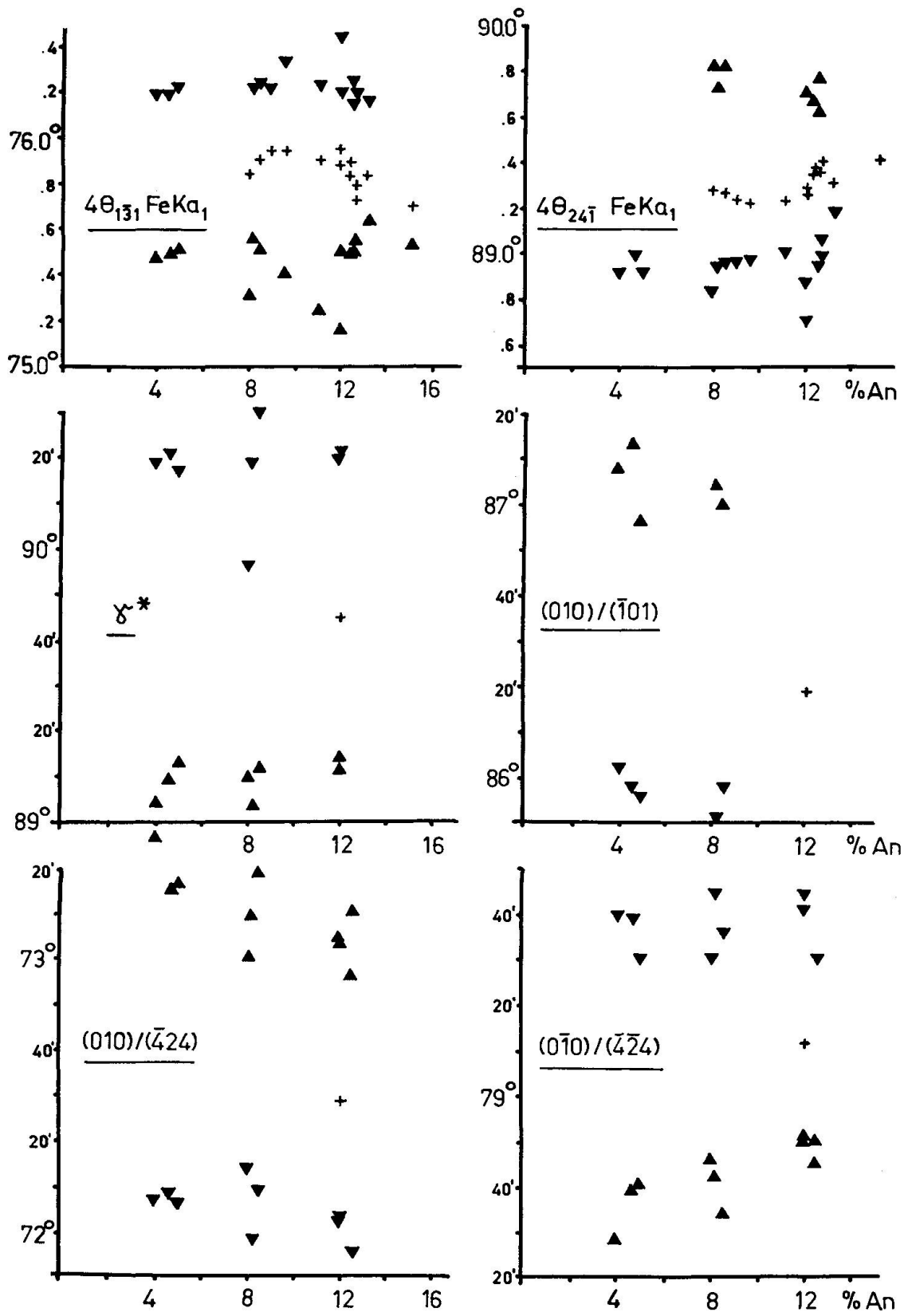


Fig. 2. Correlation of different parameters of peristerites with their bulk composition.
 ▲ = An-rich phase ▼ = Ab-rich phase + = "Third" phase

distortions of the lattices as compared with that of a pure albite (BAMBAUER et al. 1967); the two parameters c and γ show constant discrepancies.

Specimen	a	b	c	α	β	γ
172	8.142 Å	12.783 Å	7.149 Å	94.27°	116.62°	87.80°
Albite	8.140 Å	12.785 Å	7.159 Å	94.30°	116.60°	87.70°
99	8.140 Å	12.790 Å	7.148 Å	94.21°	116.63°	87.84°

If a distortion in the opposite direction is assumed for the An-rich phase there will be a compensation for the distortion of the lattice of the Ab-rich phase. It seems that the angles $010/\bar{4}24$ and $0\bar{1}0/\bar{4}24$ are not strongly affected by the distortion of these lattice constants. These parameters are, therefore, assumed to be the best ones for determining the (approximate) anorthite contents of the An-rich phases. Though it is not strictly correct to determine the An-content of the An-rich phases with the aid of the working curves given for "plagioclase (low)", it has been done because of the convincing evidence that the peristerite unmixing takes place when the plagioclase is near the ordered state.

The An-contents derived, using these parameters, are plotted against the bulk composition in Fig. 3. This diagram is very similar to that of RIBBE (1962); with an increase in the anorthite content of the bulk composition the An-content of the An-rich phase diminishes. Considering the fact that the An-contents of the An-rich phases are only approximate values, the scatter of the plotted values in this figure is considered insignificant.

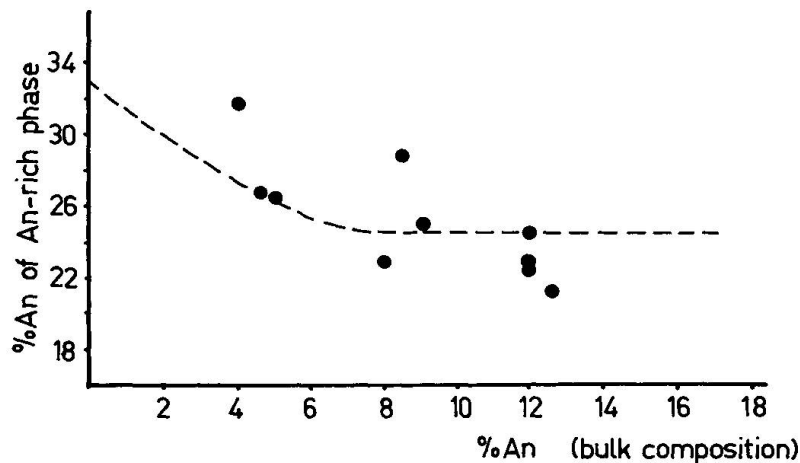


Fig. 3. Correlation of the An-contents of the An-rich phases of peristerites with their bulk composition.

It is assumed on the basis of NIGGLI's theoretical considerations that the Ab-rich phase of a peristerite is almost a pure albite. The fact that three of the calculated lattice constants of the two Ab-rich phases (a , b and β) coincide with those of a pure albite appears to support this assumption.

There is still the question of the so-called "third phase". The different parameters such as $4\theta_{(1\bar{3}1)}$ give more or less a constant value of 16% An for this phase, in spite of the differences in the bulk compositions which range from 8 to 13% An.

DISCUSSION

These observations seem to suggest that a single exsolution curve is not adequate to explain the *kinetics* of peristerite unmixing. A series of exsolution curves, each of which is characteristic of a particular composition and of a particular Al/Si-distribution appears more probable. An attempt has been made in Fig. 4 to draw a series of exsolution curves based on observed values. But with the available data we could not get any information on the temperatures of exsolution, i.e. whether the crest of the curve corresponding to An₁₆ lies higher than those of albite-rich plagioclases or vice versa.

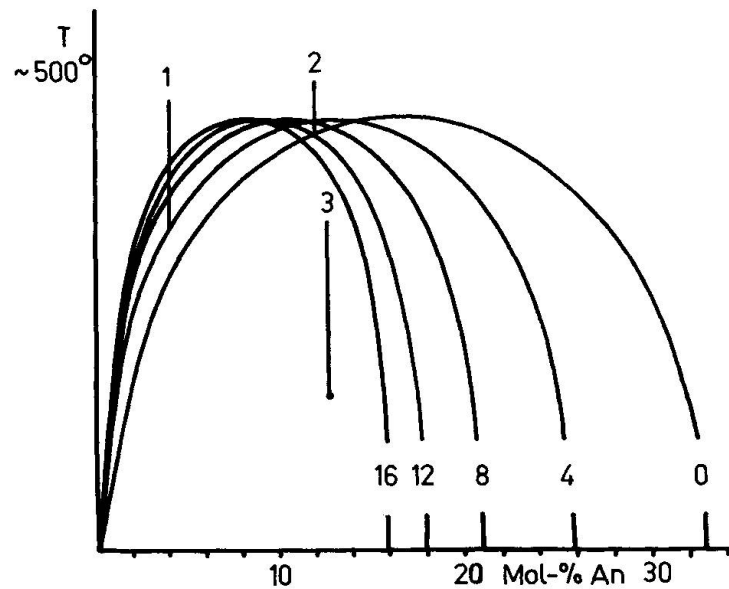


Fig. 4. A schematic diagram to illustrate the nature of exsolution of peristerites. Heights of the different curves are chosen arbitrarily. The number marked on each curve gives the characteristic bulk composition for which that particular curve is valid. See text for further details.

The assumption of a series of curves explains all the facts hitherto observed. Three cases are considered below:

Case 1: Let us suppose that a plagioclase having the composition 4% An and a stable Al/Si-distribution cools slowly from a high temperature (case 1 in Fig. 4). After sufficient cooling it will meet the exsolution curve corresponding to its composition (4% An-curve). There it will begin to unmix with an An-rich

phase containing about 22% An. During this process it becomes richer in albite. It will therefore unmix further only in accordance with the new exsolution curve corresponding to the new composition. At this second stage of exsolution another An-rich phase, richer in An-content than the first one, will be exsolved. The An-rich phases will not unmix further because crystals of composition greater than 16% An do not show any exsolution. Hence after complete unmixing there will be a pure albite phase and a series of An-rich phases varying in composition from 22% An to about 33% An (with an average value of 27% An).

Case 2: Let a plagioclase with a composition of 12% An and having a stable Al/Si-distribution cool down slowly (case 2 in Fig. 4). On reaching the corresponding exsolution curve an Ab-rich phase will be exsolved, here e.g. 6% An. As a result the original phase will become richer in anorthite. It will therefore unmix further according to the new exsolution curve corresponding to the new composition. This may go on till the original phase attains 16% An content after which it cannot unmix and hence will be present as the "third phase". On the other hand the Ab-rich phase will unmix further into albite and a series of An-rich phases varying in composition from 16% to 33%. The nearer the bulk composition of the original phase to 16% An, the greater the proportion of the 16%—phase (here called the "third phase") should be.

Case 3: Let us consider a plagioclase having a composition lying between 0% and 16% An, which for reasons stated in (b) unmixes at a temperature much lower than the equilibrium temperature corresponding to its chemical composition. This is a case of exsolution shown by a plagioclase having an unstable Al/Si-distribution. In such cases it is not possible to visualise the course of unmixing; but it is certain that the unmixing will not proceed according to any one of the curves shown in Fig. 4 (e.g. specimen No. 135). When a plagioclase of composition 8—16% An undergoes an exsolution of this type it may not be expected to show the "third phase".

The assumption of these exsolution curves explains the following observations:

1. Plagioclases having a bulk composition greater than 16% An do not show any exsolution.
2. There is no definite value for the composition of the An-rich phase and hence it always shows diffuse reflections.
3. If the bulk composition of a peristerite is greater than (appr.) 8% An, it always shows a "third phase" with a composition of about 16% An, provided it has unmixed with a stable Al/Si distribution.

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