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The origin of antiperthites in anorthosites

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Abstract

Antiperthites in anorthosites and gabbroic anorthosites from the Laramie Range and the Adirondacks apparently form by an exsolution mechanism involving nucleation and growth from original ternary feldspars containing approximately 6 percent Or. These antiperthites consist of flattened, cylindrical-shaped K-feldspar blebs of $Or_{92}An_1Ab_7$ to $Or_{96}An_{0.5}Ab_{3.5}$ occurring primarily along twin and low-angle grain boundaries of host plagioclases with $An_{40}Or_{2.5}Ab_{57.5}$ to $An_{48}Or_{2.5}Ab_{49.5}$. The following observations support an exsolution mechanism. (1) The An content of the host plagioclase increases near the K-feldspar bleb, indicating slow diffusional transport of Ca and Al exchanged for alkalies and Si away from the bleb site. (2) The flattened sides of the blebs are oriented near ($\bar{3}01$), ($0\bar{4}1$), and (100) and are subparallel to planes with minimal elastic strain in other feldspar intergrowths. (3) The crystallographic axes of the noncoherent blebs and the plagioclase host are subparallel. (4) Although homogenization of antiperthites did not occur after six months of heating at 1000°C, some Ca redistribution as well as a structural change in the host plagioclase were observed. Heating at 700°, 800°, and 900°C resulted only in redistribution of K and Na, allowing apparent tie lines between the plagioclase (An_{44}) and the alkali feldspar to be drawn. These results suggest that if temperatures are high over long periods of time, Al–Si diffuison rates are adequate to produce antiperthites by exsolution. Exsolution cannot have occurred during the crystallization of the anorthosite, since preservation of probable igneous Al–Si ordering (structural states) in host plagioclase indicates minimal Al–Si diffusion occurred during a later metamorphism.

Introduction

Antiperthites are intergrowths of K-rich alkali feldspars in plagioclase hosts with An contents commonly ranging from 0 to 48 in terrestrial samples. Antiperthites in this study are from andesine anorthosite complexes having host plagioclase compositions ranging from An_{38-48}. An understanding of the formation of these antiperthites can provide information on the crystallization temperature and cooling rates achieved during the thermal histories of anorthosites.

Three mechanisms have been postulated for the origin of antiperthites: (1) replacement of plagioclase by alkali feldspar (*e.g.*, Heier, 1955; Sen, 1959; Griffin, 1969); (2) simultaneous crystallization of an alkali feldspar and a plagioclase feldspar (Vogel *et al.*, 1968; Vogel, 1970); and (3) exsolution of an alkali feldspar from an originally homogeneous ternary feldspar. Exsolution involving nucleation and growth has been suggested from textural evidence by Carstens (1967). Exsolution involving spinodal decomposition in the early stages has been suggested by Christie (1969), Nissen *et al.* (1967) and Smith, in Smith and Ribbe (1969). Evidence for an exsolution origin, probably involving nucleation and growth, will be presented in this paper.

Antiperthites from noritic and gabbroic anorthosites in the Laramie Range and the Adirondacks were selected for detailed study. The Adirondack samples are from the St. Regis Quadrangle described by Davis (1968, 1971). The Laramie samples are from the area near the western margin of the anorthosite complex along Wyoming Highway 34, described by Skrzyniecki (1970), and from the area immediately east of there mapped by Newhouse and Hagner (1957).

Petrographic description of antiperthite

The K-feldspar in antiperthites occurs as flattened rod-shaped blebs in the host plagioclase (Fig. 1). Three bleb orientations can sometimes be seen in a single host grain, and combinations of differing orientations are often responsible for irregularities of

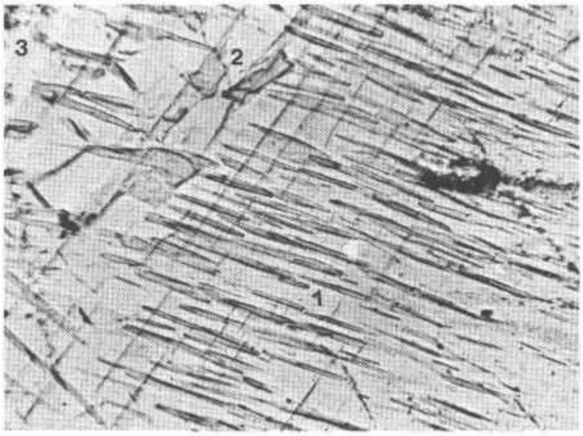

Fig. 1. Photomicrograph showing K-feldspar blebs in a plagioclase host from a Laramie Range gabbroic anorthosite (LR-17). The shape of the blebs can be determined by observing blebs in the three orientations (×475), labeled 1, 2, 3 on the photomicrograph.

shape seen in some blebs. For example, Z-shaped blebs probably result from the combination of two orientations. Separate blebs that apparently grew in different orientations terminate against each other rather than cross-cut each other. Slight variations of orientation are often observed within a set of blebs oriented in the same general direction. Most blebs are not randomly distributed, but tend to be concentrated and evenly developed along imperfections such as twin and low-angle grain boundaries in the host plagioclases. Other blebs, not as obviously associated with imperfections, may be related to submicroscopic features such as twins and dislocations.

Irregular-shaped grains or blebs of K-feldspar are also found at grain boundaries in more noritic or gabbroic samples. These blebs may or may not be associated with the neighboring plagioclase. The frequency of marginal blebs, as well as their size and shape irregularities, increases with the mafic mineral content of the rock.

Chemical description

Chemical analyses were performed using an automated ARL electron microprobe at Brown University. The electron beam was focused to one μm, and analyses were made by stepping in two μm intervals across the plagioclase host and the K-feldspar blebs. Since most blebs were four to six μm across, only one or two points could be analyzed within a bleb.

Host plagioclases with An_{40-47} contained abundant K-feldspar blebs, whereas those with greater than 48 percent An had few K-feldspar blebs, but often contained Bøggild lamellae (fine-scale sub-microscopic intergrowth of two plagioclase phases seen with a JEM-7A TEM). A few samples around An_{48} had both antiperthites and Bøggild lamellae. Plagioclases with An-rich cores (48) and An-poor rims (42–44) were antiperthitic only at the rims. The Or content of all host plagioclases ranged from 2.5 to 3.5. Compositions of the K-feldspar blebs ranged from Or_{94-97} and had approximately An_1.

The nature of compositional zoning associated with the occurrence of the K-feldspar belbs is illustrated by two traverses perpendicular to the long dimensions of the blebs. The first traverse shown in Figure 2 is across one K-feldspar bleb and the surrounding plagioclase. The composition of the host away from the bleb is uniformly $An_{44}Or_3Ab_{53}$, and the composition of the bleb is $Or_{95.5}Ab_{3.5}An_1$. Approximately ten μm to the left of the bleb the host An content begins to increase, reaching a maximum of An_{49} adjacent to the bleb. This value must be regarded as a minimum, due to limitations resulting

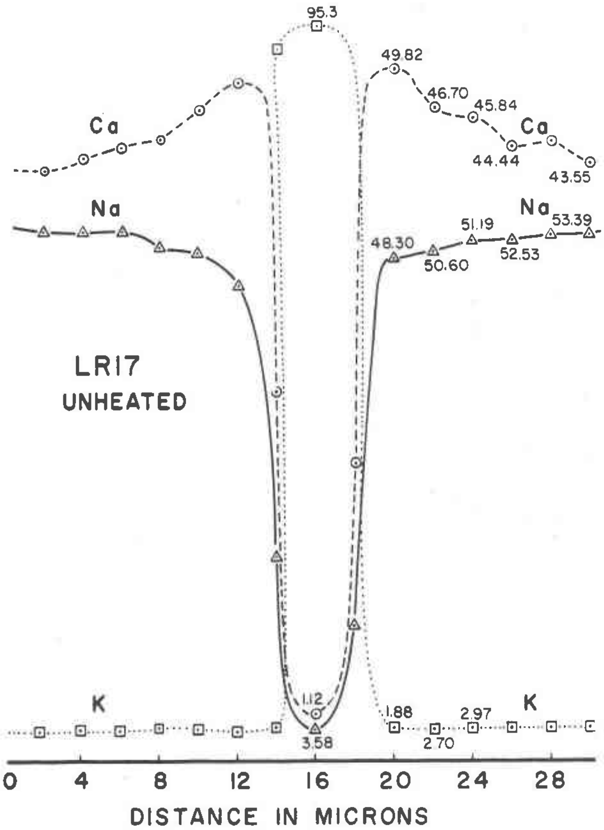

Fig. 2. Electron microprobe traverse showing change in composition as a function of distance across a K-feldspar bleb in sample LR-17. Numbers on traverse are mole percent An on Ca line, Ab on Na line, and Or on K line.

from the resolution of the electron beam relative to the scale of the zoning. The Or content of the host plagioclase appears to remain relatively constant, decreasing only slightly where the An content is the highest. The zoning pattern on the right side of the grain is almost symmetrical to the left side. The second traverse, across the three blebs and the host in the same sample, is shown in Figure 3, and illustrates that the increase is host An content near a bleb depends on the bleb size and the proximity of other blebs.

A mass-balance calculation was made to determine whether the excess An content around the bleb margins was sufficient to offset the deficiency within the blebs. As an example, the bleb in Figure 2 was modeled as an elliptical cylinder with a 10 μm major diameter and a 4 μm minor diameter. An average increase of 0.42 weight percent Ca over the host value of 6.50 weight percent was assumed to occur over a 10 μm distance on all sides of the bleb. The model accounts for 110 percent of the required An. Considering possible errors due to approximations in the model, the mass-balance calculation appears to justify the assumption that the excess An accounts for the rearrangement of Ca, Al, and Si necessary to exsolve the antiperthite from a homogeneous ternary feldspar.

Finally, analyses were done to determine the composition of the ternary feldspar needed to produce the present antiperthites by exsolution. Potassium X-ray distribution photographs were taken on the electron microprobe for a wide variety of bleb orientations and distributions. The fractional area covered by the blebs varied from 0.71 to 8.23 percent and averaged 3.88 percent. Using the compositions of the blebs and the host in Figure 2 and assuming the blebs constitute 3.88 percent of the total volume, the approximate initial composition for the homogeneous ternary feldspar would be $An_{44}Or_6Ab_{50}$. Ternary feldspars of this composition have been found in volcanic rocks, and permit an exsolution origin.

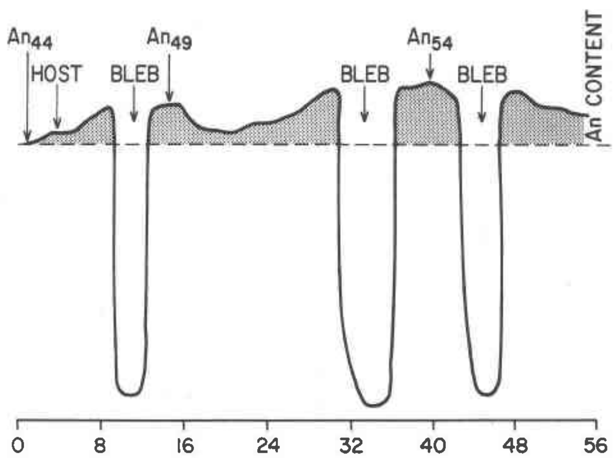

Fig. 3. Electron microprobe traverse across three K-feldspar blebs showing change in weight percent An as a function of distance in sample LR-17. Hachured area above dashed line represents excess An surrounding the blebs.

Orientation of the blebs

Reports of antiperthite orientation in the literature have consisted of flat-stage measurements of the angular relationship between the long dimension of blebs and cleavage or twin planes in the host (Naidu, 1954; Sen, 1959). These data are incomplete and unsatisfactory for comparison with orientations of other feldspar intergrowths.

The orientations of the flattened surface of the blebs, measured on a five-axis universal stage in this study, are given in Figure 4. Since the area of the flat surface is small, and the blebs occur on discontinuities, measurements relative to a uniform part of the host are difficult to make and errors are large. Usually, only measurement of one of the three orientations which may occur in a single grain is better than approximate. Some scatter is real, and reflects a small range of variation within a single orientation.

Antiperthite orientations can be compared to observed and theoretically calculated orientations for other feldspar intergrowths. Williame and Brown (1972, 1974) have calculated interfacial energy of all possible orientations for perthites, peristerites, Bøggild and Huttenlocker lamellae using elastic constants and cell parameters, and have found minimum energy orientations that compare favorably to measured orientations of naturally occurring intergrowths. The contours for the interfacial energy of peristerites, which have the same basic shape as for the other plagioclase intergrowths, are shown superimposed on the antiperthite orientation diagram in Figure 4. The two areas of lowest interfacial energy and hence most probable orientations for exsolution correspond to the two largest clusters of antiperthite data points. Although it is difficult to separate real data scatter from measurement error, the similarity of the antiperthite orientations to other feldspar intergrowths thought to form by exsolution is consistent with an exsolution origin for antiperthites. The variability of the antiperthite orientation may reflect the greater differences in cell parameters between K-feldspars and plagioclases than between two plagioclases. Since the flattened blebs are not parallel to

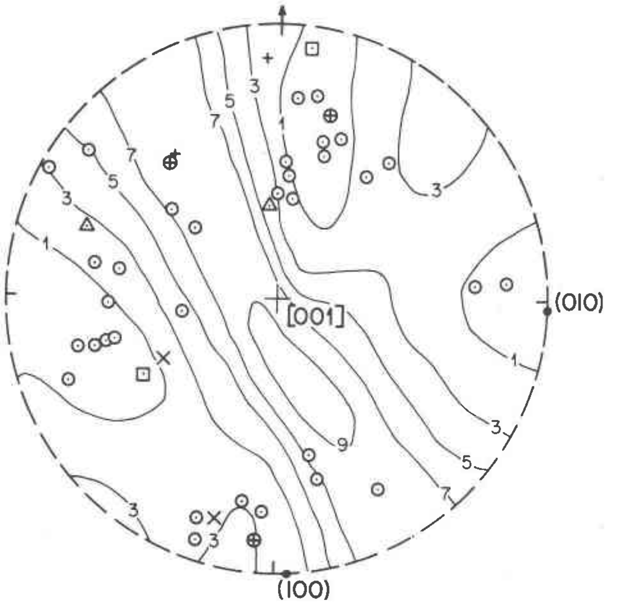

Fig. 4. Wulff-net stereographic projection of poles to the planes of the flat surface of the K-feldspar blebs. Open circles represent grains where only one orientation was measured. Other symbols represent grains where more than one orientation was measured. Similar symbols are for multiple measurements within the same grain. The superimposed contours are from Willaime and Brown (1972) and show the probability of coherent exsolution in different orientations for peristerites. The most probable orientations are within the 1 contours.

Poles to the orientations of K-feldspar blebs were measured on a five-axis univeral stage. The measurements were made relative to the optic axis of the plagioclase host and were rotated to the crystallographic axis of an An_{45} plagioclase on the stereonet. The crystallographic axis of the An_{45} plagioclase was determined by interpolating between the values given for An_{40} and An_{50} by Slemmons (1962).

prominent growth facies, a criterion suggested by Smith (1974b), the orientation data do not support a simultaneous crystallization origin.

X-ray analyses

Single-crystal X-ray precession photographs were made of a number of antiperthitic grains. All plagioclases have low to low-transitional structural states, based on the α^*–γ^* angles and the presence of "*e*" reflections. A second set of "*a*" reflections corresponding to the K-feldspar blebs occurs subparallel to a^*, b^*, and c^* of the plagioclase, but is most easily seen along a^* because of the large difference in the *a* cell dimension between plagioclase and K-feldspar. The presence of two sets of "*a*" reflections indicates that the two phases are non-coherent. Two K-feldspar maxima, three and four degrees apart, are seen in some samples and are probably related to more than one orientation of K-feldspar blebs. Similar diffraction features were observed by Bown and Gay (1971) in lunar antiperthites and by Bown and Gay (in Smith, 1974a) in an An_{50} antiperthitic plagioclase from Essex County, New York. A similar explanation for double maxima was presented by Bown and Gay (1971).

Potassium–feldspar structural states were determined by measuring the difference in spacing between the ($\bar{2}$01) peak of K-feldspar and the (101) peak of KBr on powder diffraction patterns (Orville, 1963). The structural state of the K-feldspar appears to be variable and to correlate with the size of the blebs. Larger blebs in Adirondack samples tend to have lower structural states than the smaller blebs in Laramie samples.

The structural state of the plagioclase was determined using the (131–1$\bar{3}$1) peak separation method of Bambauer *et al.* (1967). Generally, chemical composition must be known before structural state can be determined. However, the low-structural-state plagioclases between An_{38-48} have a nearly constant value of (131–1$\bar{3}$1) (Bambauer *et al.*, 1967). The (131–1$\bar{3}$1) peak separation thus provides a measure of structural state for plagioclases in this study. The Or component in the plagioclase also affects the (131–1$\bar{3}$1) spacing, but can be neglected when comparing relative changes in this study, since all plagioclases contain $Or_{2.5}$ to $Or_{3.5}$.

Both Laramie and Adirondack anorthosites contain plagioclases with low to low-transitional structural states. Variations within a single hand specimen appear to be nearly as large as variations across the field areas studied (see Table 1). The total range is greater in Adirondack samples than in Laramie Range samples (see Table 1). Detailed study shows that generally the lowest structural states are found in the phenocryst cores and the highest structural states are found on the rims and in small grains.

An attempt was made to correlate structural-state variation with antiperthite bleb density. The smear slides that were scanned for (131–1$\bar{3}$1) peak separation were also scanned for the presence and intensity of the ($\bar{2}$01) alkali feldspar peak. No correlation between the intensity of the ($\bar{2}$01) peak and the degree of ordering of the plagioclase was found, suggesting that the presence or absence of an antiperthitic intergrowth is not the determining factor in the structural-state variation in the plagioclase.

This conclusion is in agreement with the findings of Romey (1969), who attributed differences in the structural state of plagioclase within hand specimens

Table 1. Representative (131-1$\bar{3}$1) spacings for host plagioclases (gabbroic anorthosites)

A. Range in Single Hand Specimen (7 separate slides each)

Laramie Range LR-17	Adirondacks SR-5
1.746	1.745 (core)
1.747	1.748
1.747	1.786 (core)
1.757	1.797
1.731	1.809
1.763	1.821
1.765	1.832 (rim)

B. Plagioclase Structural State Compared with Intensity of ($\bar{2}$01) Peak in Alkali Feldspars

Sample	(131-1$\bar{3}$1)	($\bar{2}$01)
LR-9		
large phenocryst	1.719	none
large phenocryst	1.725	none
groundmass	1.788	strong
groundmass	1.821	weak
SR-3		
phenocryst	1.748	weak
phenocryst	1.739	none
groundmass	1.791	weak
groundmass	1.815	moderate

of some Adirondack anorthosites to local differences in the igneous crystallization histories. If Romey's interpretation is correct, the role of metamorphism in antiperthite formation is probably minor. If the metamorphism that the Adirondack anorthosite has suffered (Buddington, 1939) did not alter the structural states of the plagioclases, then Al–Si diffusion must have been minimal, and the large degree of Al–Si rearrangement necessary to form antiperthites by exsolution could not have occurred at this time. The antiperthites must, therefore, have exsolved at high temperatures during the crystallization of the anorthosite.

Results of heating experiments on the antiperthites

Heating experiments were conducted on antiperthitic samples to determine the extent of antiperthite homogenization possible at laboratory times and temperatures. Small blocks of the sample wrapped in platinum foil were heated dry at 100°C intervals from 700° to 1200°C and one atmosphere for times ranging from one week to six months. Compositional changes were determined for both the host and the blebs by electron microprobe analyses. Plagioclase structural state changes were monitored by examining the (131–1$\bar{3}$1) peak separation. In addition, single-crystal precession photographs were taken at each temperature for a sample successively heated for two weeks each at 950°, 1050°, and 1100°C and one atmosphere.

Little or no change was observed in the Ca–Al content of the K-feldspar blebs in runs below 1000°C. As expected from self-diffusion coefficients in alkali feldspars (Foland, 1974; Kasper, 1974), K–Na exchange between the host and the blebs occurred readily, and apparent equilibrium was reached with respect to Na and K in the alkali feldspars after ten weeks at 700°, between five and ten weeks at 800°, and in five weeks at 900°C. No change in K content was detectable in the host because of the large volume of host relative to the blebs. Apparent tie lines for the K-feldspar blebs and plagioclase host are shown at 700°, 800°, and 900°C in Figure 5.

At 1000°C, some coupled exchange of Ca and Al from the host for alkalis and Si in the bleb was inferred from increases of Ca and Al in the blebs. Na–K exchange was very pronounced. Figure 6 illustrates the increase in Ca in the blebs, as well as the larger concentration change of Na relative to Ca due to more rapid K–Na diffuison rates. Since Ca–Al and alkali–Si coupling are necessary to maintain charge balance, the slow Ca–alkali exchange rate reflects the difficulty of Al–Si diffusion. In ten weeks at 1000°C, the average An content of the blebs changed from 1.1 to 2.6 and the Ab content changed from $Ab_{4.5}$ to $Ab_{42.5}$, and apparent Na–K equilibrium was reached. The apparent tie line is shown in Figure 5. Since attainment of apparent Na–K equilibrium requires a longer time at 1000° than at 900°C, additional move-

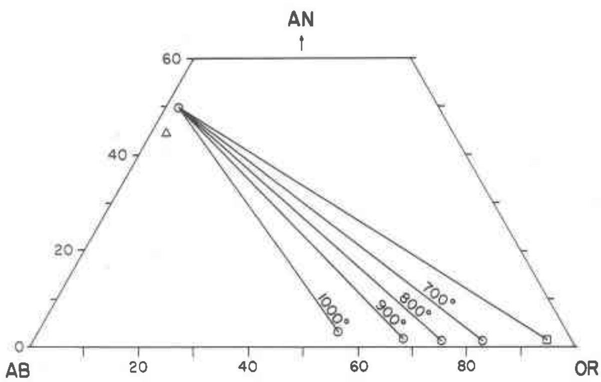

Fig. 5. Apparent tie lines between the alkali feldspar blebs and host plagioclase in sample LR-17 determined experimentally by heating in air at one atmosphere. Tie lines are drawn to An-rich plagioclase surrounding alkali feldspar blebs. The open triangle represents the general host plagioclase composition. The open square represents the original composition of the unheated blebs.

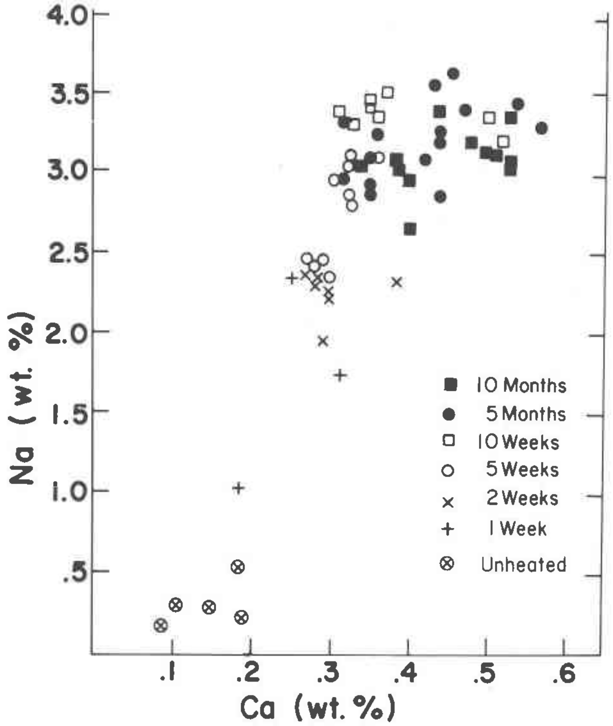

Fig. 6. Plot of weight percent Ca versus weight percent Na for alkali feldspar blebs in sample LR-17 unheated, and heated dry at 1000°C for one, two, five and ten weeks, and five, and ten months.

ment of Ca and Al apparently retards the K–Na exchange rate. Microprobe data were not sensitive enough to detect An changes in the gradients surrounding the K-feldspar blebs.

Since the alkali-feldspar blebs melted in the 1100°C runs, nothing was learned about homogenization rates at higher temperatures. Melting apparently occurs around 1060°C, as indicated on the binary phase diagram for the alkali feldspars at one atmosphere.

Changes in "*e*" reflections were observed during step heatings of a single crystal of LR-17. After two weeks, the reflections were largely unaffected at 950°, became increasingly diffuse at 1000° and 1050°, and were either too diffuse to observe or not present at 1100°C. Similar behavior of "*e*" reflections with heating has been related to changes in structural state by Bown and Gay (1958). As shown in Figure 7, measurement of (131–1$\bar{3}$1) peak separations in X-ray powder patterns of heated samples also indicated that observable disorder did not occur until 1000°C. Significant disorder was observed after longer heating times at 1000° and in shorter heating times at 1050°, 1100°, and 1200°C. Both powder patterns and single-crystal precession photographs indicate that measurable Al–Si diffusion does not occur until 1000°C in dry experiments, and are consistent with the observed increase in An in the K-feldspar blebs at 1000°C.

The "*a*" reflections in the K-feldspar blebs became increasingly diffuse until they were barely visible at 1050°C. These reflections were absent at 1100°C due to melting. Changes in the spacing of the reflections along the a^* axis with increasing temperature can be approximately correlated with the increase of sodium in the blebs. Estimates of bleb composition can be obtained from a^* using the data of Orville (1963).

The results of the heating study suggest that, at high enough temperatures over long periods of time, diffusion rates of Al and Si are adequate to produce antiperthites by exsolution. Apparent equilibrium of the noncoherent blebs with the host at temperatures near the melting point of alkali feldspar suggests that the chemical solvus lies above this temperature and that exsolution probably occurred at a coherent or semicoherent solvus. The lack of homogenization at high temperatures may result from failure to reestablish coherency of the blebs and the host (see Yund and McAllister, 1970).

Summary and conclusions: an exsolution model for antiperthites

A model for antiperthite formation from ternary feldspars in anorthosites by an exsolution process involving nucleation and growth is suggested, and is based on the observations on natural samples as well

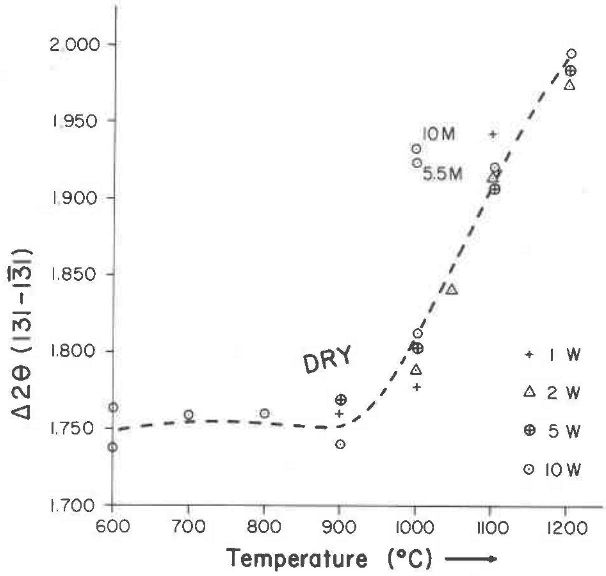

Fig. 7. Change in the spacing of (131–1$\bar{3}$1) as a function of temperature for the host plagioclase in LR-17 after heating from 600° to 1200°C. Cu$K\alpha$ radiation.

as the experimental evidence. Ternary feldspar begins crystallizing in an anorthosite complex at temperatures between 1000° and 1100°C (Anderson, 1968; Anderson and Morin, 1968; Taylor, 1968). As the temperature drops, the feldspar composition passes below the solvus associated with the ternary miscibility gap and the K-feldspar nucleates. The nucleus might be initially coherent or semi-coherent, as suggested by the greater similarity of cell parameters for the two phases at higher temperatures (Grundy and Brown, 1967, 1972), the near parallellism of the crystallographic axes, and the similarity of antiperthite orientations to other feldspar intergrowths thought to exsolve coherently. However, any coherency is probably lost at a very early stage, since the difference in cell parameters is great enough to result in a large strain for a coherent nucleus. This large strain is apparently minimized by nucleation on host imperfections. The initial composition of the nucleus is probably controlled by the stable tie lines at the exsolution temperature. As exsolution proceeds, Al and Ca are expelled from the bleb area and, due to the slow rate of Al–Si diffusion, gradients develop on the margins of the blebs. If, as the temperature drops, the Al and Si become immobilized before the An gradients are equilibrated with the host, the gradients are preserved. Meanwhile, Na–K diffusion continues and the tie lines rotate towards the Or corner of the feldspar triangle with decreasing temperature. The final position of the tie lines defines the "quenching-in" temperature of the antiperthites.

Since Al–Si diffusion appears to have been minimal during a postulated later metamorphic event in the Adirondacks, a probable lower limit for the temperature and pressure of antiperthite exsolution corresponds to the metamorphic conditions. The metamorphism is in the garnet–clinopyroxene "subfacies" of the granulite facies which has an estimated temperature of 800°–900°C and a pressure of 8–10 kbar (Martingnole and Schrijver, 1971, based on the work of Green and Hibberson, 1970).

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