Termination of the 2 feldspar + liquid curve in the system Ab-Or-An-H₂O at low H₂O contents

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ABSTRACT

Analysis of the termination topology of the plagioclase + alkali feldspar + liquid (PAL) curve in the system Ab-Or-An(-H₂O) was conducted using thermodynamic solution models for ternary feldspar melt and solid solutions for a low activity of H₂O and a pressure of 2 kbar. Under these conditions the PAL curve terminates within the ternary system and does not lie within a thermal valley near its termination. Rather, the adjacent liquidus surfaces show a peritectic-like topology with the lowest temperatures lying on the Ab-Or(-H₂O) sideline. The calculated PAL curve has an almost constant slope to its termination; this is in disagreement with the geometric conclusion of Henderson (1984). Using Henderson's geometric method supplemented by an analytical approach, we show that no sharp decrease in An is required as the termination is approached.

Behavior of bulk compositions close to and on the line that connects the consolute point of the solvus (K) with the melt composition at the termination (C) of the PAL curve has been investigated computationally. Bulk compositions slightly displaced from the K-C line will undergo complete resorption of one of the feldspars before the solidus is reached. However, compositions on the K-C line will show coprecipitation of two feldspars during equilibrium crystallization with the subtraction of a crystalline component from each of the solids, a process that we term "partial resorption." Partial resorption removes the mass-balance requirement that the PAL boundary must curve away from K.

INTRODUCTION

The system Ab-Or-An(-H₂O) (i.e., the feldspar system) (Fig. 1) represents a model system from which insights can be obtained into the crystallization paths of feldspars in more complex multicomponent systems. Phase relations in this system are difficult to determine experimentally because of sluggish reaction kinetics between plagioclase and melt (Johannes, 1979). However, the data at 5 kbar of Yoder et al. (1957) in this system saturated with H₂O as well as the data of Tuttle and Bowen (1958) in the system Ab-Or-Qz-H₂O indicate the presence of a two feldspar + L (PAL) curve in the An-poor region of the system. The common use of the term "cotectic" for this curve is misleading because, as recognized by Tuttle and Bowen (1958), crystallization along the curve may be either even or odd. Ricci (1966) termed the former eutectic-like or coprecipitational and the latter peritectic-like or resoritional. Therefore, in this article this curve will be referred to as the PAL curve (plagioclase, alkali feldspar, and liquid).

Recent work by Nekvasil (1990) uses a thermodynamic approach to supplement the sparse experimental data in the feldspar system and to evaluate the effects of pressure and activity of H₂O (a₂) on the relative position of the PAL curve and the corresponding section of the ternary feldspar solvus. The calculated phase relations were used to determine the compositional regions in which bulk compositions must lie for complete or incomplete resorption of one of the two feldspars to take place. The termination of the PAL curve and its topologic relation to the consolute point on the solvus section was not considered in great detail by Nekvasil (1990). However, the topology of this region has been the object of intense geometric analysis by Stewart and Roseboom (1962) and Henderson (1984) and figures prominently in the evaluation of fractional crystallization paths conducted by Abbott (1978).

Stewart and Roseboom (1962) considered the possible terminations of the PAL curve depending upon the relative positions of the consolute point, the termination of the PAL curve, and the nature of the three-phase triangles. Henderson (1984) applied an unpublished graphical construction developed by J. W. Greig to evaluate the geometry of the region close to the termination using hypothetical three-phase triangles and a hypothetical consolute point. He concluded that geometry required that the PAL curve be the tangent to the line K-C at the termination; therefore, the PAL curve must turn sharply to lower An contents near the termination. This article evaluates the topology in this region through calculation of the phase equilibria using thermodynamic mixing models for ternary feldspar solid solutions (Lindsley and Nekvasil, 1989) and feldspathic melts (Burnham and Nekvasil, 1986; Nekvasil, 1988a). This approach comple-
ments the geometric approach and permits quantitative evaluation of the required geometry.

THE AN-POOR REGION OF THE TERNARY FELDSPAR SYSTEM

Phase relations in the region around the termination of the PAL curve were calculated using the revised quasi-crystalline model of Burnham and Nekvasil (1986) for the thermodynamic properties of the melt in the system Ab-Or-An-H$_2$O and the ternary feldspar model of Lindsley and Nekvasil (1989). Detailed comparisons of calculated phase equilibria against available experimental data have been discussed in Nekvasil and Burnham (1987) and Nekvasil (1988a, 1988b). The phase equilibria calculations were conducted as described in Nekvasil (1988a) with a few modifications. At the given pressure and activity of H$_2$O, a search routine was used in order to determine the composition of melt and the temperature at which a three-phase assemblage was stable. Because of the extraordinary sensitivity of the crystalline phase compositions to the melt composition in the region near the PAL termination, together with the relative insensitivity of the slope of the solvus tie lines to temperature, calculations of the melt-feldspar equilibrium were used only to deduce the activities of the crystalline components and the equilibrium temperature. The compositions of the feldspars were then obtained by matching the crystalline component activities to the sixth decimal place using the activity matching routine for solvus tie lines applied by Nekvasil and Burnham (1987).

The general topology of the albitic region of the ternary feldspar system and the crystallization trends are relatively insensitive to the solid and melt models used in the calculations, although the exact positions and equilibration temperatures of the PAL curve and the solvus section are sensitive to the solid-solution model chosen. This was demonstrated by using the melt model of Burnham and Nekvasil (1986), an ideal melt model, and the solid-solution models for ternary feldspar of Nekvasil and Burnham (1987), Fuhrman and Lindsley (1988), and Lindsley and Nekvasil (1989). Thus, we believe the general topologic relations presented here to be correct, although phase compositions and equilibrium temperatures will vary depending upon choice of models. The discussion below focuses on the calculated results based on the melt model of Burnham and Nekvasil (1986) and the ternary feldspar solution model of Lindsley and Nekvasil (1989).

The investigation of the geometric relations in the region of the termination of the PAL curve was conducted for low H$_2$O activity, as this results in the greatest separation of the termination from the Ab-Or-(H$_2$O) sideline (Nekvasil, 1990). Figures 1 and 2 show the calculated liquidus surface in the albitic region of the ternary feldspar system at 5 kbar and H$_2$O-saturated conditions and at 2 kbar and a$_{H_2O}$ = 0.1, respectively. In Figure 1, the PAL curve is even (eutectic-like) along its entire extent and intersects the Ab-Or sideline in a eutectic. In contrast, the PAL curve becomes odd at a specific point in Figure 2, and its peritectic-like characteristics are indicated by the configuration of the A + L isotherms that are convex toward Or, as opposed to those in Figure 1 that are concave toward Or. It must be noted that the PAL curve becomes peritectic-like at a unique point. However, this point does not necessarily indicate the melt composition at which coprecipitation is replaced by resorptions behavior for equilibrium crystallization (Nekvasil, 1990). In the peritectic region, the liquidus temperatures in the bounding subsystem Ab-Or-(H$_2$O) are lower than those along the PAL curve, and melts leaving this curve are not constrained to follow a thermal valley to the An-free sideline. For higher H$_2$O contents, the transition between the topology for a eutectic (thermal valley) and the topology for a peritectic of the Af + L surface lies at increasingly higher Af contents. Because of the peritectic topology in this system at high temperatures, incomplete or complete resorption of a feldspar is expected at some stage of the equilibrium crystallization history for melt compositions reaching the odd region of the PAL curve.

Figure 3a shows several calculated three-phase triangles along the albitic portion of the PAL curve for a$_{H_2O}$ = 0.1 and 2 kbar. (Additional three-phase triangles were also calculated and used in construction of the geometric elements but have been omitted from Fig. 3a for clarity.) In the region near the termination of the PAL curve, very slight changes in temperature and hence liquid compo-
Fig. 3. Detailed geometry of plagioclase (P), alkali feldspar (A), and melt (L) compositions (in mol%) in the vicinity of the termination (C) of the PAL curve. K is the consolute point on the polythermal solvus section (curve P₁-K-A₁) corresponding to the temperatures along the PAL curve. The curves TSC (which represents the compositions of the total solids), P + L, and A + L (which are the compositions of plagioclase plus liquid and alkali feldspar plus liquid, respectively) reflect variations in the phases during equilibrium crystallization along the PAL boundary. Arrows indicate the direction of decreasing temperature. (a) Relations calculated for 2 kbar and $a_0 = 0.1$, as outlined in the text. Some of the three-phase triangles used in the calculation of the TSC, P + L, and A + L curves have been omitted for clarity. Note that the calculated PAL curve shows little curvature. The instantaneous solid compositions (ISC), as defined in the text, are $λ_{1,5}$ through $λ_{11,5}$. Calculated temperatures (°C): $T_1 = 1076.77$; $T_2 = 1074.97$; $T_3 = 1074.12$; $T_4 = 1073.24$; $T_5 = 1072.8$; $T_6 = 1072.54$; $T_7 = 1072.45$. (b) Relations deduced, mainly on geometric grounds, by Henderson (1984). Compositions are schematic only. Note the inferred strong curvature of the PAL boundary away from K near its termination C.

sition result in marked changes in the compositions of coexisting feldspars. The feldspar vertices of the three-phase triangles converge toward the consolute point K ($An_{18},Ab_{30,5}Or_{31,0}$), becoming increasingly narrow until they reach the limit described by the K-C line (Henderson, 1984; Nekvasil, 1990). For conditions of equilibrium crystallization, melts can reach the termination C ($An_{17},Ab_{37,46}Or_{38,77}$) and feldspars the consolute point K only if the bulk composition lies on the tie line K-C. Bulk compositions outside of the solvus section, yet within the envelope described by the three-phase triangles on the plagioclase side of the K-C line, must undergo complete resorption of alkali feldspar during equilibrium crystallization before the solidus is reached; those on the alkali
feldspar side will undergo complete resorption of plagioclase. The variation of phase proportions during crystallization is critically dependent upon the geometry of the three-phase triangles, which in turn are dependent upon the relative configuration of the solvus section in the region of K and the PAL curve near C.

**Geometric Constraints on the Termination Topology**

Using an unpublished geometric method developed by Greig, Henderson (1984) investigated the geometric relations of the solvus section and the PAL curve near C. He concluded that the PAL curve must move sharply to lower An compositions as C is approached (Fig. 3b). Such a geometry departs significantly from the calculated geometry shown in Figure 3a. In order to understand the differences between the calculated diagrams and those constructed from geometrical considerations, we apply the method used by Henderson (1984) but use our calculated (rather than Henderson’s estimated) three-phase triangles and a chosen bulk composition I along the K-C line.

Following the method of Henderson (1984), we can project lines from each vertex of the three-phase triangles in Figure 3a through bulk composition I to obtain the proportions of the other two phases. For example, a line from liquid 1 through I intersects the plagioclase–alkali-feldspar side of the three-phase triangle I at the ratio of the two feldspars. Likewise, a line from the plagioclase vertex through I intersects the liquid–alkali-feldspar side at the ratio of those two phases. However, as Henderson pointed out, the geometric construction becomes increasingly difficult as the triangles become narrower (i.e., as the two feldspar compositions approach K). For this reason, we have determined the intersections analytically rather than graphically. Using the calculated compositions of the phases for each three-phase triangle and bulk composition (I), we solved for the intersection of two lines: the line from a vertex through I and the opposite side of the triangle. For a given bulk composition, those intersections for a succession of triangles define three curves: the total solid composition (TSC), the alkali feldspar + liquid (A + L), and plagioclase + liquid (P + L) curves (Fig. 3a) for equilibrium crystallization. As P and A converge toward K and L approaches C, the P + L and the A + L curves each approach a limit. This limit is not the composition I because the ratio K1 to IC indicates the proportions of a single feldspar (the sum of the proportions of A and P). Furthermore, this limit is the same for each of the curves only if the same amounts of A and P were crystallizing per unit change of the melt along the PAL curve. For any composition I along the K-C line, more A than P crystallizes per unit change of the melt composition near the termination, and the A + L and P + L curves do not converge.

Based on his graphical constructions, Henderson (1984) adduced two lines of evidence to support his conclusion that the PAL curve, near its termination, must move tangentially away from K. One of his arguments—that the A + L and P + L curves must converge upon C (Henderson, 1984; his Fig. 36c, p. 75)—has been withdrawn as erroneous (Henderson, written communication, 1989). His second argument is based on mass balance; if Ab, An, and Or components are being added to each of the two precipitating feldspars from the melt as the two co-precipitating feldspars approach K (and thus each other) the liquid path (i.e., the PAL curve) must move away from K. We show below, however, that in the region near the termination, precipitation of two feldspars does not necessarily imply that all three components being subtracted from the liquid are added to both of the feldspars.

**Variations in Instantaneous Transfer Compositions**

**Bulk compositions on the K-C line**

That the calculated PAL boundary does not curve still seems to defy petrologic intuition: if the feldspars are changing composition by addition of material from the melt, must not the liquid move away from K (Henderson, 1984)? The answer to this question lies in the evaluation of the instantaneous transfer compositions, which are the composition of the material being subtracted from the liquid (z of Henderson), that of the material added to plagioclase (π), and that of the material added to alkali feldspar (α) at any instant. The instantaneous melt transfer composition, λ (equivalent to the instantaneous solid composition, ISC), is given by the intersection of the tangent to the curve describing the variation in bulk compositions of the solid (TSC of Fig. 3a) and the back tangent to the liquid curve at any instant. The intersections of the tangents to the TSC and the back tangents to the liquid curves can be obtained geometrically. However we have obtained them analytically by approximating the tangents using the line segment between two adjacent three-phase triangles. The intersections of the tangents approximated in this manner (e.g., using the line segments between triangles 1 and 2) yield λ1, through λ115, shown in Figure 3a. These transfer compositions would be required to approach the K-C line if the melt were to move abruptly toward the Ab-Or sideline as suggested by Henderson (1984).

Inasmuch as the position of the instantaneous liquid transfer composition is dependent upon the transfer compositions of the solids, the reason for the location of the values of λ as shown can perhaps be best illustrated by analyzing the positions of the instantaneous plagioclase (π) and alkali feldspar (α) transfer compositions. As indicated by Henderson (1984), π is given by the intersection of the tangent to the plagioclase composition and the tangent to the A + L curve at any three-phase triangle. Similarly, α is given by the intersection of the tangent to the alkali-feldspar composition and the tangent to the P + L curve at any three-phase triangle. These intersections are exceedingly difficult to obtain geometrically, however, because of the very acute nature of the three-phase tri-
angles, the curvatures of the A + L and P + L curves, and the near parallelism of some of the tangents near the termination. Therefore, they were obtained analytically in this study. Figure 4 indicates the position of several \( \pi \), \( \alpha \), and \( \lambda \) values using the approximation described above. The instantaneous alkali-feldspar transfer compositions (\( \alpha \)) are positive with respect to all three components (with the exception of \( \alpha_{1,3} \)), and, because they lie in the prograde direction from A, they indicate that alkali feldspar is being precipitated. Likewise, the \( \pi \) compositions show that plagioclase is also being precipitated. However, the material added to plagioclase (\( \pi \)) is negative in An content (i.e., it lies at negative An values). This means that An component is being removed from P even as Ab and Or components are being added.

The intersection of the line \( \alpha-\pi \) with the tangent to the TSC also gives \( \lambda \) without use of the L curve; the locus of \( \lambda \) points obtained in this manner is coincident with those obtained from the liquid. It is the negative An content of \( \pi \) that exerts a major influence on the position of the ISC (\( \lambda \)) and indicates that the melt will not simply move away from the solid composition as the consolute point is approached.

The portions of the total mass of the system that are present in each phase during equilibrium crystallization were calculated to investigate further the crystallization behavior of compositions in which crystallization of plagioclase occurs through the addition of Ab and Or components and the subtraction of An component. The calculated three-phase triangles were used to compute the variation in phase proportions during equilibrium crystallization along the PAL curve for the bulk composition \( I (Ab_313,An_{39},Or_{31}) \) (Fig. 5a) that lies on the calculated K-C line. Equilibrium crystallization of this bulk composition shows coprecipitation behavior for both feldspars (Fig. 5a), that is, neither feldspar undergoes complete or incomplete resorption before the solidus is reached. The fraction of the system that consists of each component in each phase can be readily computed for one mole of material by multiplying the phase proportions (e.g., \( X_p \)) by the composition of each phase (e.g., \( X_p' \)). The difference between any two points is a measure of the amount of each component being added to the bulk plagioclase, or alkali feldspar, or subtracted from the liquid. Such computations for the well-known plagioclase melting interval show that although the mole fraction of An in plagioclase decreases with decreasing temperature, the total mass of An in bulk plagioclase increases. The negative \( \pi \) values in Figure 4 indicate a different behavior. Whereas the proportions of plagioclase and alkali feldspar increase throughout the crystallization interval considered, the total amount of An component in plagioclase decreases over the entire interval, as shown by the An curve in Figure 5b. (Figure 5 is based on the mole number of grams of An in plagioclase decreases over this interval. This is in sharp contrast to the binary plagioclase system in which the mole fraction, but not the mass, of An in plagioclase decreases by dilution during crystallization. An in the ternary plagioclase here is being removed, not simply diluted.) The Or component in alkali feldspar (Fig. 5c) shows a similar behavior, but only close to the termination, while the melt composition is varying smoothly (Fig. 5d). The phenomenon of decreasing An or Or during precipitation of a feldspar (and hence, a negative value of any one component in \( \pi \) or \( \alpha \)) we term “partial resorption.” Partial resorption is a result of more rapid decrease in the An content of plagioclase or Or content of alkali feldspar than increase in amount of the phase precipitating. This occurs only in the region of the solvus characterized by major changes in the plagioclase and alkali-feldspar compositions (i.e., around the nose of the solvus). Partial resorption removes the mass-balance requirement that the PAL boundary near C must cur be away from K. This is because the total mass of feldspar can change composition both by addition of material and by subtraction of a component. Thus, the PAL curve of Figure 3a does not violate mass-balance constraints.

**Bulk compositions off of the K-C line**

Compositions lying near the K-C line, but to the plagioclase side of it, will undergo complete resorption of alkali feldspar before the liquid can reach C during equilibrium crystallization (Henderson, 1984; Nekvasil, 1990). Figures 6a–6d show the variation in phase proportions as well as the masses of each component in the feldspars for the bulk composition \( Ab_{33.3},An_{33.3},Or_{33.3} \), which lies close to I, but on the P + L side of the last three-phase triangle in Figure 3a. During equilibrium crystallization, the amount of plagioclase increases throughout the crystallization interval, slowly at first and then more rapidly as alkali feldspar is being resorbed (Fig. 6b). However,
Fig. 5. Calculated phase proportions and variations in composition during equilibrium crystallization along the PAL curve (increasing Ab content of the liquid) for bulk composition I (Ab$_{53.33}$An$_{13.00}$Or$_{33.67}$), which lies on the K-C line (Fig. 3a). (a) Variations of phase proportions. The numbers shown refer to the corresponding three-phase triangles in Figure 3a. The proportions of both plagioclase and alkali feldspar increase throughout the crystallization interval. (b) Variations of component abundance in plagioclase (expressed as mol% of the total system). A decrease corresponds to a decrease in the total mass of a component in the plagioclase, regardless of whether the mole fraction of that component or the phase proportion of plagioclase is increasing or decreasing. The decrease in An with decreasing temperature, therefore, means that the An component is being removed (partially resorbed) even while the total amount of plagioclase is increasing. (c) Variations of component abundance in alkali feldspar (expressed as mol% of the total system). Note that there is a small amount of partial resorption of Or component near the the PAL termination C. (d) Variations of component abundance in liquid (expressed as mol% of the total system).

the mass of An in total plagioclase initially decreases because the amount of plagioclase initially increases more slowly than the decrease in An content of the plagioclase; thus initially plagioclase is undergoing partial resorption. However, as the resorption of alkali feldspar commences and abundant An is being released back into the melt, the increase in the proportion of plagioclase during crystallization is more rapid, and partial resorption is replaced by conventional precipitational behavior. During equilibrium crystallization, the amount of alkali feldspar first increases and then decreases to zero, indicating first coprecipitational behavior and then resorptional behavior (Fig. 6a). The mass of each component in the total alkali feldspar mimics this trend by first increasing and then decreasing (Fig. 6c). It is interesting to note that the variation in the components in the melt phase does not
Fig. 6. Phase proportions and variations in component abundances during equilibrium crystallization along the PAL curve (increasing Ab content of the liquid) of the bulk composition Ab$_{33.33}$An$_{13.69}$Or$_{53.63}$, which lies near I but on the plagioclase side of the K-C line (Fig. 3a). (a) Variations of phase proportions. The numbers shown refer to the corresponding three-phase triangles in Figure 3a. Note the initial gradual increase in abundance of Af as coprecipitational behavior is manifested and then the abrupt decrease in Af as resorptonal behavior commences and proceeds to completion. (b) Variations of component abundance in plagioclase (expressed as mol% of the total system). Note that the An component undergoes partial resorption until the late stages, when alkali feldspar begins to resorb and the proportion of plagioclase increases dramatically. (c) Variations of component abundance in alkali feldspar (expressed as mol% of the total system). Abundances of all components increase early in the crystallization sequence, then decrease as alkali feldspar begins resorbing. (d) Variations of component abundance in liquid (expressed as mol% of the total system).

reflect the change from coprecipitational behavior with partial resorption of An from plagioclase to complete resorption of alkali feldspar (Fig. 6d). Figures 7a–d indicate the analogous case for composition Ab$_{52.71}$An$_{15.63}$Or$_{34.29}$, which lies close the K-C line but on the alkali feldspar side. Alkali feldspar increases throughout the interval, most notably when plagioclase is being strongly resorbed. As plagioclase is resorbed throughout the crystallization interval along the PAL boundary, no partial resorption behavior is manifested.

CONCLUSIONS

From the above analysis, we have shown that there is no requirement for the PAL curve to curve sharply to more An-poor compositions as its termination is approached. Rather, the almost constant slope of the cal-
Fig. 7. Phase proportions and variations in component abundances during equilibrium crystallization along the PAL curve (increasing Ab content of the liquid) of the bulk composition Ab$_{32.32}$An$_{13.80}$Or$_{54.38}$, which lies near I but on the alkali-feldspar side of the K-C line (Fig. 3a). (a) Variations of phase proportions. The numbers shown refer to the corresponding three-phase triangles in Figure 3a. Plagioclase abundance does not vary much initially but drops to 0 as the liquid reaches L$_{12}$ (Fig. 3a). (b) Variations of component abundance in plagioclase (expressed as mol% of the total system). Note that the An component is being resorbed throughout the crystallization path along the PAL boundary. Or increases initially, then decreases abruptly along with Ab and An as plagioclase is being resorbed. (c) Variations of component abundance in alkali feldspar (expressed as mol% of the total system). The abundances of all components increase throughout the crystallization path along the PAL boundary sequence. (d) Variations of component abundance in liquid (expressed as mol% of the total system).

culated PAL boundary is a more reasonable representation of the geometry of the true PAL curve. However, as the compositions of the solids change around the nose of the solvus, the solid composition added to either feldspar may include a negative amount of An or Or. This partial resorption phenomenon strongly affects the instantaneous solid composition and negates the requirement that the melt move away from the feldspar composition as the consolute point is approached. Compositions along the K-C line will not undergo complete resorption of either phase but will show partial resorption of An in plagioclase and Or in alkali feldspar as the termination is approached. The equilibrium crystallization behavior discussed for compositions along and to either side of the K-C line will describe crystallization of albitic compositions as long as the termination of the PAL curve lies within the feldspar system and has not reached the Ab-Or(-H$_2$O) boundary. The calculated topology upon which
these conclusions were based is not sensitive to the ther-
modynamic models chosen. The same shape of the PAL
curve and its relation to the solvus was reached for sev-
eral melt and solid-solution models, indicating that this
behavior is a necessary part of crystallization in the ter-
nary feldspar system at high temperatures.

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