## Calorimetric and phase-diagram approaches to two-feldspar geothermometry: a critique

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## Abstract

Two-feldspar geothermometers based on the thermodynamic properties of the limiting binaries Ab-An and Ab-Or are unsatisfactory because the ternary nature of the feldspar pairs cannot be accounted for and critical points are not generated. Ternary mixing data are mandatory. There are also serious shortcomings in the experimental ternary phase relation data available at present, and even ambiguities in the interpretation of the thermodynamic properties of the binaries, which preclude development of a practical geothermometer. Recent work on feldspar zoning patterns in rocks allows some deductions to be made concerning the geometry of the ternary system, but there are difficulties in the interpretation of natural feldspar pairs. Framework order-disorder presents problems which may be insuperable.

#### Introduction

The problem of constructing a two-feldspar geothermometer has been tackled in two ways, using on the one hand a thermodynamic approach based on calorimetric and cell-volume data, and on the other a direct experimental phase synthesis approach. Both approaches attempt to depict the same relations in the ternary feldspar system, and both approaches have severe experimental problems. However, as used up to the present, only the thermodynamic approach has serious theoretical shortcomings as well. These were discussed by Brown and Parsons (1981). However, because new thermochemical data have recently been presented by Haselton et al. (1983), and used to construct a revised two-feldspar thermometer using the earlier approach, we here reluctantly reiterate our objections to the theory, and also make further comments (and suggest some experiments) on the interpretation of the available thermodynamic and phase synthesis data, on the relationships in natural feldspars and on the problem of order in the feldspars.

# "Double-binary" thermometers for disordered feldspars

Haselton et al. (1983) have recently presented carefully determined low-temperature heat-capacity data on a range of disordered binary alkali feldspars, and provided a comprehensive review of other thermodynamic data on alkali feldspars. They use their new data in conjunction with calorimetric results on plagioclases synthesized at 1200°C obtained by Newton et al. (1980), to construct a modified version of the two-feldspar thermometer presented by Stormer (1975) and Powell and Powell (1977). The new ther-0003-004X/85/0304-0356\$02.00 mometer is expressed both as a formula and in graphical form.

Their calculated isotherms are reproduced here in Figure 1B. Like its predecessors, the thermometer is based on the supposition that the ternary feldspar system can be treated as two independent binaries, but the Haselton et al. version differs from earlier versions in treating high plagioclases as non-ideal (using the Newton et al., 1980 data) and in using their own new revised mixing parameters for the alkali feldspar binary.

Brown and Parsons (1981) showed that the doublebinary assumption is erroneous and leads to a thermometer which conflicts both in general form and in detail with established ternary phase relationships, irrespective of the quality of the data on the two binary systems. In view of the credibility lent to the thermometer by the authors of the recent paper, and by the appearance of a version of such a thermometer in a recent textbook (Ehlers and Blatt, 1980), it seems necessary to repeat our reasons for considering that the thermometer is impractical in its present form.

1. The fundamental misconception embodied in the double-binary thermometers lies in the way the minor components are treated. The activity of Ab is considered to be affected neither by Or in plagioclase nor by An in alkali feldspar. This can be true only if the solubility and therefore the Henry's law coefficients for the minor components are nearly the same for all compositions in the binary. However, the activity coefficient of An in alkali feldspar must vary from nearly unity in albite (with which it mixes nearly ideally, see below) to a very large number in sanidine (with which mixing is extremely limited). The effect of adding small amounts of a minor component to Ab-poor



Fig. 1. Comparison of a two-feldspar geothermometer based on ternary phase synthesis data (A) with one based on the thermodynamic mixing properties of the two binaries Ab–Or, and Ab–An (B).  $N_{Ab,PL}$  and  $N_{Ab,AF}$  are mole fractions of Ab in plagioclase and alkali feldspar respectively. (A) Isotherms from Seck (1971a) at 1 kbar, except 910°C, 0.5 kbar. The binary solvus and its critical point  $T_c$ , are from Smith and Parsons (1974). S' and S" are alternative ternary critical points corresponding with the alternative interpretation of the critical solution curve suggested in the present Fig. 3. The temperature of 1060°C suggested for the isotherm on which they lie, is from Parsons and Brown (1983). While we stress that some of the data on which this diagram is based may have low accuracy, the thermometer does have the correct general form. (B) Isotherms based on the "double binary" approach as given by Haselton et al., 1983. However good the thermodynamic data for the binaries, the thermometer departs markedly from the correct form particularly at T > 800°C, and isotherms also extend into the "forbidden" region corresponding with the binary alkali feldspar solvus.

feldspars may be decisive. For example, an An-free alkali feldspar Or<sub>73</sub>Ab<sub>27</sub>An<sub>0</sub> (Y, Fig. 2a) will be homogeneous at 800°C (Smith and Parsons, 1974), but substituting only 3 percent An for Or (Z) will give a two-feldspar assemblage (Seck, 1971), with a plagioclase Or<sub>6</sub>Ab<sub>50</sub>An<sub>44</sub>(Z'). The double-binary thermometers predict that the same An-free alkali feldspar can be in equilibrium with a range of plagioclases up to Or<sub>0</sub>Ab<sub>47</sub>An<sub>53</sub>, at a range of T between 500 and 1000°C (Fig. 2b). Adding only about 3% An to this alkali feldspar, in place of Or, giving Or70Ab27An3, strikingly changes both the equilibrium tie-lines, and the temperatures. It is clear that the "minor" components strongly perturb the activity-composition surface. All equilibrium feldspar pairs must lie on the ternary solvus and are thus saturated in all components. The minor components cannot be treated as non-participating impurities. They are, in the present context, in no sense minor, but are critical to the application of the ternary thermometer.

A further shortcoming of the double binary thermometer, which results from the omission of the minor components, is that the ternary compositions of natural feldspars must be projected onto the binary joins in some more or less arbitrary way. Haselton et al. adopted the formulation of Powell and Powell (1977) which is one of several means of projecting the ternary compositions onto the binary joins. They project Or in plagioclase along lines of constant Ab onto the plagioclase join (S'on Fig. 2a) and An in alkali feldspar is omitted, by projecting parallel to the plagioclase join (P, Fig. 2a) for  $N_{\text{Or,AF}}$  and parallel to the anorthitesanidine join for  $N_{Ab,AF}$  (S), the mole fractions respectively of Or and Ab in alkali feldspar. In effect, this implies that Or has the same effect on the mixing properties of plagioclases as does An, and that An can substitute with equal effect for Ab and Or in alkali feldspars. This is most unlikely.

2. When the minor components have been dealt with in the above way, it is possible to calculate isotherms which show the compositions of coexisting pairs of feldspars. Such isotherms must not extend into regions of temperature-composition space which are forbidden by the phase relations: this includes almost the whole of the area to the right of the line marked "binary solvus" in Figure 1A. As can be seen, the Haselton et al. curves extend into this region and would all terminate at a point where  $N_{Ab,AF} = N_{Ab,PL} = 1$ .

In fact isotherms may terminate in only one of two ways. First, for T below that of the critical temperature  $(T_{\rm crit})$  of the binary alkali feldspar solvus, isotherms *must* terminate on the binary solvus. Such isotherms at T not far below  $T_{\rm crit}$  (Fig. 2a) may cross the projected solvus and then terminate on it (e.g., the 650°C isotherm, Fig. 1). This is because such isotherms have maxima in  $N_{\rm Ab,PL}$  before they reach the Ab–Or join. Second, isotherms at  $T > T_{\rm crit}$  must terminate on the  $N_{\rm Ab,PL}/N_{\rm Ab,AF} = K_{\rm D} = 1$  line. This is because all solvus isotherms above the binary  $T_{\rm crit}$  have their own critical points, the ternary locus of which is the *critical* solution curve (Fig. 3). The exact shape of this curve is not well known and evidence bearing on its location is dis



Fig. 2. Ternary phase diagrams for the system Or-Ab-An. (a) Solvus isotherms at 650 and 900°C from Seck (1971) at 1 kbar and 0.5 kbar respectively. HT is the limit of feldspar solid solution suggested by Parsons and Brown (1983), which probably corresponds with a high-temperature isotherm at  $\sim 1060^{\circ}$ C. Z-Z' is a tie-line at 800°C, 1 kbar, from Seck (1971), Y-Y' is an An-free feldspar pair at 500°C, 1 kbar, from Smith and Parsons (1974). The arrows, P, S and S', illustrate methods of projecting ternary compositions onto the binary joins. (b) Plagioclases predicted to be in equilibrium with composition Y on the Haselton et al. model at various temperatures.

cussed below. However, isotherms must terminate on the  $K_{\rm D} = 1$  line at compositions near to  $N_{\rm crit}$  of the binary solvus, and move towards lower values of  $N_{\rm Ab}$  at higher temperatures (Fig. 1).

The presence of critical points on solvus isotherms above  $T_{\rm crit}$  is an essential feature of the ternary feldspar system which is not accommodated in the double-binary model. It was a feature fully appreciated by Seck (1971) whose thermometer was based on ternary phase relations, discussed below. We modified his thermometer in this respect as he showed all isotherms terminating at one value of  $N_{\rm Ab}$ , whereas we think it most likely that  $N_{\rm Ab}$  varies with T in the general manner shown, a point also discussed by Seck.

We, therefore, conclude that a useful approximation to a feldspar geothermometer cannot be obtained by combining the thermodynamic properties of the binary joins, however excellent the measurement of those properties. *Ternary* mixing parameters must be obtained first. A useful thermometer should furthermore be formulated in such a way that the likelihood that the phases are in equilibrium can be assessed, and there is no more elegant device for this than the ternary phase diagram. In the next section we discuss the present state of knowledge on the geometry of the ternary feldspar system, and some of the problems of the attainment of equilibrium in natural systems.

## Ternary feldspar phase relationships

#### Experimental studies

The experimental determination of the ternary feldspar phase relationships is difficult for two main reasons. One concerns the compositions of the phases, and is instrumental, the other concerns the attainment of equilibrium, and is kinetic.

Composition of the feldspar phases. As the crystals produced in direct crystallization experiments are generally too small for chemical analysis by electron microprobe methods, an indirect method has hitherto been used. The solubilities of An in alkali feldspar and of Or in plagioclase are only up to about five percent in Ab-poor members, so the high precision required is difficult to attain. All methods used are graphical and rely heavily on the determination of the Or-content in either phase from the 201-spacing. Two different procedures have been used. In the first, the limit to the one-feldspar field is determined separately and is then used to determine the compositions of coexisting pairs using a construction through the bulk composition (Hamilton, 1969; Seck, 1971; Norris, 1972). In the second, two separate bulk compositions on a possible tie-lie are used to fix the compositions of coexisting pairs and hence the limit to the one-feldspar field (Seck, 1971; Smith, 1978). The precision of either method is not high. The compositions of the starting gels are probably known to a precision of about 1-2% of the feldspar components and the Or-contents of the coexisting pairs to a precision of



Fig. 3. Aspects of the ternary system deduced from natural assemblages. Zoning patterns in two sets of natural feldspars are shown to illustrate the problems in interpretation. Areas labelled N are for a nepheline trachyte lava (Carmichael et al. 1974), triangles and dots are respectively for continuously zoned plagioclase and alkali feldspar crystals in a teschenite sill (Henderson and Gibb, 1983). Areas M and C are respectively mesoperthite and cryptoperthite in a syenogabbro, believed to have crystallized as separate homogeneous phases (Parsons and Brown, 1983; Brown and Parsons, 1983). S-S' and S-S'' are alternative critical solution curves consistent with the two zoning patterns. S, the critical composition on the Ab-Or binary, is from Smith and Parsons (1974). T is the limit of feldspar solid solution suggested by Tuttle and Bowen (1958), P that suggested by Parsons and Brown (1983).

about 2% at the best, depending on the quality of the diffraction pattern of the run products and on their state of order. It is not surprising that small discrepancies exist between the reported results. It is thus indispensible to develop new methods for the direct determination of phase compositions, or for growing larger and more homogeneous crystals.

Problems of exchange equilibrium and kinetics. It was shown by Parsons (1978) that in most of the experimental work on the binary Ab–Or solvus, equilibrium was not attained. Most of the published so-called solvi are not solvi at all but simply curves showing compositions of feldspar pairs, both of which were changing composition with time in the same direction. This situation was explained on theoretical grounds by Smith and Parsons (1974) and Parsons (1978). Such curves should not be used in thermodynamic treatments of the binary system although they frequently are (see for example, Haselton et al., 1983).

Similar problems of non-equilibrium must exist in the ternary system and would show up in kinetic studies. In the studies of Seck (1971) and Norris (1972), data were not given concerning change of composition with time nor were the compositions of coexisting pairs bracketed from both directions. It is not possible to affirm that the published results correspond to equilibrium, but, if experience in the binary system where there is no problem with Al/Si exchange can be used as a guide, this seems most improbable. The only study in which both kinetic data were given and brackets were achieved is that of Smith (1974, 1978) for compositions with only up to 10% An. His results do not show the very impressive regularity of those of Seck (1971) or Norris (1972). This regularity does not necessarily imply a good approach to equilibrium, by analogy with the behavior on the Ab-Or binary (Smith and Parsons, 1974). Similar partial equilibrium is detectable in the data of Seck (1972) on the Ab-Or binary (see discussion in Parsons, 1978), but such effects cannot be deduced from his data in the ternary system. We believe there is an urgent need to determine the directional and kinetic aspects of equilibration in crystallizing gels or glasses of ternary feldspar composition. Johannes (1979) has carefully investigated the rate of equilibration between limited pairs of feldspars, using the Viswanathan (1971) ion-exchange method to obtain albite contents of plagioclase, and has suggested that equilibrium is unlikely to have been reached in Seck's (1971) experiments. He provided two rather different tielines at 800°C.

In conclusion, we have serious doubts as to whether the ternary data of Seck (1971) represent complete equilibrium (metastable, however, with respect to Al–Si order–disorder) particularly when the difficulties experienced by other workers are taken into account. The apparent regularity of the data may possibly reflect partial exchange equilibrium in which both phases are changing composition with time, as discussed by Parsons (1978). The system should be restudied taking careful account of the kinetics of equilibriation.

#### Al–Si order-disorder

Alkali exchange equilibrium may well occur between entirely disordered feldspars at the time of growth. Experimental synthesis studies of ordering in albite (e.g., Mac-Kenzie 1957; Mason, 1979) always produce disordered crystals at first which subsequently approach a supposed equilibrium degree of order for the temperature of synthesis. There are no experimental data on equilibrium degrees of order in K-feldspars, calcic plagioclases or any ternary composition, but it is highly likely that in nature some degree of ordering may occur immediately subsequent to crystallization, which may or may not be accompanied by redistribution of Ab, Or and An between the feldspar phases. In long-term isothermal exchange experiments the degree of order will increase with time and approach the complete stable equilibrium values, which will be different in the two phases. Any future experimental study should address this problem. As we have noted previously (Brown and Parsons, 1981) in natural examples of ordered feldspar pairs it is difficult to know at what stage, if ever, alkali exchange equilibrium is reached, and it is probably best to assume disorder at the time of alkali exchange.

Although in alkali feldspars ordering may be continuous, this is not the case in basic plagioclases, which may undergo a  $C1 \rightleftharpoons I1$  phase transition. The equilibrium temperature of this transition depends on composition in the Ab-An system (Carpenter and McConnell, 1984), but the effect of Or is unknown. Even natural plagioclases may well crystallize with the  $C\overline{1}$  structure, as such plagioclases frequently show b-antiphase boundaries (see, for example, Ribbe, 1983) which must have been produced on ordering after growth. A TEM study of natural Or-bearing basic plagioclases coexisting with alkali feldspar, would be of great interest in this respect. The presence of b-APBs in such feldspars would indicate that deviations from ideality in Ab-An mixing due to this transition (see Carpenter and Ferry, 1984) should be ignored and only the C1 structure taken into account when formulating a thermometer.

Haselton et al. (1983) attempted to improve on earlier thermometers (e.g., Stormer, 1975) by using the Newton et al. (1980) high temperature solution calorimetry. This they interpreted as showing that departures from ideality in high plagioclases are predominantly at the Ab-rich end of the plagioclase series (Haselton et al., 1983, Fig. 7). Carpenter and Ferry (1984), however, reinterpreting the same calorimetric data in the light of the  $C\overline{1} - I\overline{1}$  transition, suggest that departures from ideality are at the An-rich end of the series, and that  $C\overline{1}$  plagioclases can be considered to show ideal mixing. It is apparent that the thermodynamic data base for a two-feldspar geothermometer is fraught with problems different in type, but equally as hard to solve as those posed by phase-synthesis data.

## Natural assemblages

The critical solution curve. Some aspects of the ternary system may be deduced by considering the compositions of natural feldspars. Of the many studies of zoning patterns, particularly useful examples are provided by Carmichael et al. (1974) and Henderson and Gibb (1983), shown on Figure 3. Carmichael et al. studied groundmass feldspars in a nepheline trachyte lava, while the Henderson and Gibb examples are continuously zoned crystals in a teschenite sill. In principle, feldspar pairs crystallizing in such rocks should lie either on isotherms or intersect them slightly down-temperature. Provided that the two feldspars are growing together and are instantaneously in equilibrium, the critical composition on the ternary solvus must lie between them. The locus of critical compositions on different isotherms defines the critical solution curve of which possible traces, SS' and SS" are shown on Figure 3. Different magma types will generate different solidus-solvus intersections because they have different solidus temperatures, but the critical solution curve on the ternary solvus is independent of melt composition since it depends only on the partitioning of Ab, Or and An between the two feldspar phases, provided no other components enter the feldspar.

Interpretation of natural zoning patterns is not unambiguous, as our examples show. Curves SS' and SS" (Fig. 3), are respectively consistent with the zoning patterns of the nepheline-trachyte and the teschenite. In the teschenite, Henderson and Gibb (1983) showed continuous zoning of plagioclase and alkali feldspar crystals meeting at a composition of An<sub>3</sub>Ab<sub>55</sub>Or<sub>42</sub>. However, the alkali feldspar phase in the nepheline-trachyte appears to zone continuously past this composition; there is a substantial gap between the most Ab-rich alkali feldspar and the most Or-rich plagioclase, through which the alternative critical curve, SS', has been drawn (see Parsons and Brown, 1983). A possible explanation of such conflicting relationships (suggested by C. M. B. Henderson, pers. comm., 1983) is that one feldspar phase (in patterns like that of the nephelinetrachyte) is in reaction relationship with liquid (rather than coprecipitation relationship), and this may generate an apparent gap in zoning trends which does not necessarily straddle the critical solution curve. A recent treatment of the complex relationships near the termination of the twofeldspar boundary curve on the liquidus in Or-Ab-An is given by Abbott (1978).

The type of textural detail which may be needed to unravel these relationships can be illustrated by the transmission electron microscope (TEM) and microprobe (EPMA) studies of Parsons and Brown (1983) and Brown and Parsons (1983). In these studies we showed that two feldspars with bulk compositions M and C (Fig. 3) grew in contact on the strain-free ternary solvus (and therefore must straddle the critical solution curve) and subsequently exsolved largely by spinodal decomposition at the ternary coherent spinodal.

We suggest that careful combined TEM/EPMA studies are needed to reconcile different zoning patterns. However, whatever its exact path, the critical curve must ultimately turn towards the An–Or join, implying that high-T isotherms on a graphical thermometer (Fig. 1) must terminate on the  $K_D = 1$  line at progressively lower values of  $N_{Ab}$ . It is important to know the exact location of the critical curve because feldspar phases at equilibrium (which are therefore potentially useful for geothermometry) *must* straddle it.

Limits of solid solution. Limits of solid solution in the ternary system can be set by considering the bulk composition of both homogeneous and exsolved feldspars, provided that intergrowths can be shown to have formed by exsolution rather than by other processes such as simultaneous growth. It may be possible for single-phase feldspars to grow metastably inside the strain-free solvus, and even inside the coherent spinodal, but it seems highly improbable that two feldspars could grow simultaneously from melt except on the strain-free ternary solvus. Pairs such as M and C (Fig. 3) are therefore particularly informative. Bulk composition M lies well inside the two-feldspar field (T) given by Tuttle and Bowen (1958) and Parsons and Brown (1983) have suggested the new limit of solid solution (P) shown. Feldspars of composition M are mesoperthites with largely coherent lamellar intergrowths (shown by TEM, Brown and Parsons, 1983), with great regularity of lamellar periodicity over large volumes, which is quite independent of crystal boundaries (Brown and Parsons, 1983). These features strongly suggest exsolution by spinodal decomposition of a homogeneous high-temperature ternary feldspar. It would be extremely interesting to investigate other strongly ternary feldspars in this way, possibly expanding further the one-feldspar field in the ternary system. High-temperature, low-pressure rocks are the likely setting for such feldspars, because the effect of P on the solvus is greater than that on the dry melting curves for end-member feldspars (see Brown and Parsons, 1981, p. 370).

Isotherms. As we note above, zoning patterns in groundmass feldspars or those forming from residual liquids in strongly fractionating magmas should lie on isotherms or intersect them slightly down-temperature. It is striking that many zoning patterns (e.g. many in Carmichael et al., 1974, Henderson and Gibb, 1983, including both shown on Fig. 3, and Baldridge et al., 1983) cut obliquely up-temperature, across the isotherms of Seck (1971). This strongly suggests that the latter approach the Ab-corner too closely and is a major reason why further experimental work in this ternary system is required.

#### Conclusions

We reiterate our previous assertion (Brown and Parsons, 1981) that the theroetical shortcomings of the "double binary" thermometer, based on thermodynamic data, are such as to render it impractical. Nothing is to be gained by using ever more refined thermodynamic data for the binaries because the thermometer generated by this approach has an incorrect general form (Fig. 1). Ternary mixing parameters are required to generate a practical thermometer, and at the present time we have serious doubts as to whether meaningful mixing parameters could be extracted from existing ternary phase synthesis data. Controversy exists concerning the interpretation of calorimetric data for the binaries, and no such data exist for ternary feldspars. Even if a precise analytical formulation of the mixing properties could be obtained we suggest that the ternary phase diagram, and its derivatives, have overwhelming practical advantages in handling real mineral compositions. The great sensitivity of the thermometer to the "minor" components, Or in plagioclase, and An in alkali feldspar, the kinetics of Ca-Na-K exchange (and necessarily coupled Al-Si exchange), and the effect of framework order, will always provide serious problems of interpretation in natural feldspar suites.

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