

Liquidus phase relationships in the system KAlSi_3O_8 - $\text{CaAl}_2\text{Si}_2\text{O}_8$ - KAlSiO_4 at $P(\text{H}_2\text{O}) = 5$ kbar

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ABSTRACT

Liquidus phase relationships in the system KAlSi_3O_8 - $\text{CaAl}_2\text{Si}_2\text{O}_8$ - KAlSiO_4 (Or-An-Ks) have been determined for low anorthite contents (<20 wt% An) at a pressure of H_2O of 5 kbar. A temperature maximum occurs on the plagioclase + leucite field boundary, and two ternary eutectics have been defined: the assemblage plagioclase + kalsilite + leucite + liquid at 735 °C at the composition $\text{Q}_{21}\text{An}_{14}\text{Ks}_{65}$, and the assemblage alkali feldspar + plagioclase + leucite + liquid at the composition $\text{Q}_{28}\text{An}_8\text{Ks}_{64}$, at a temperature slightly higher than 735 °C. Fractionation trends in the high-K, silica-undersaturated rocks from the Roccamonfina complex of the Roman Volcanic Region, Italy, may be partially explained by phase relationships in the system Q-An-Ks at $P(\text{H}_2\text{O})$ on the order of 5 kbar, or at a higher total pressure and H_2O -undersaturated conditions. However, Na-bearing normative components in these rocks are also important to the interpretation of fractionation trends.

INTRODUCTION

K-rich, silica-undersaturated igneous rocks are observed in only limited occurrences throughout the world, and the origin of their unusual chemistries is a matter of debate. A knowledge of phase relationships in systems relevant to these rock types may help to interpret differentiation and crystallization processes within suites of K-rich lavas such as those found in the Roman Volcanic Region, Italy. Several authors have suggested that the volcanic series in this region represent suites of lavas related by fractional crystallization. For example, Appleton (1972) studied rocks from the Roccamonfina volcano and suggested that significant fractionation took place under a pressure high enough to reduce in extent the stability field of leucite. Cundari (1975, 1979) and Cundari and Mattias (1974) suggested that fractionation had also taken place in the Sabatini and Vico regions, but at lower pressures where leucite fractionation is possible.

The system SiO_2 - $\text{CaAl}_2\text{Si}_2\text{O}_8$ - NaAlSiO_4 - KAlSiO_4 (Q-An-Ne-Ks) is applicable to late-stage differentiation and crystallization in K-rich rock suites. Phase relationships in the silica-undersaturated part of the system Q-Ne-Ks have been studied extensively, but the effect of the addition of an anorthite component to this system is not well known. The experimental study we report here describes liquidus phase relationships for ternary compositions in the system KAlSi_3O_8 - $\text{CaAl}_2\text{Si}_2\text{O}_8$ - KAlSiO_4 (Or-An-Ks) at $P(\text{H}_2\text{O}) = 5$ kbar. Previous experimental studies pertinent to the system at a pressure of H_2O of 5

kbar include data along the binary join An-Or obtained by Yoder et al. (1957) as part of a study of the ternary feldspar system, and data along the binary join Or-Ks determined by Zeng and MacKenzie (1984) as part of a study of the ternary system Q-Ne-Ks. Jones and MacKenzie (1989) describe phase relationships in Na-bearing compositions in the system Ab-Or-An-Ne-Ks.

EXPERIMENTAL PROCEDURE

Experiments were carried out in an internally heated pressure vessel at a pressure of 5.0 ± 0.2 kbar. Pressure was measured using a manganin cell, and temperature was measured with two Pt/Pt 13% Rh thermocouples situated at either end of the sample holder. The temperature gradient within the sample holder was <2 °C, except in one experiment in which sample temperatures were not well constrained and were between 735 and 795 °C. Temperature was calibrated against the melting point of NaCl. Approximately 300 mg of starting material was sealed in a Pt capsule with sufficient deionized H_2O added to ensure that the sample was H_2O saturated (>13 wt% H_2O). Duration of the experiments was 4 d for all the data reported. Samples were quenched from temperatures of 735–790 °C to lower than 300 °C in about 2 min. Products were examined optically and analyzed by X-ray diffraction, using $\text{CuK}\alpha$ radiation. Electron microprobe analyses were obtained using a Cameca Camebax microprobe at an accelerating voltage of 15 kV, using a Link systems energy-dispersive spectrometer. Back-scattered electron imaging was used to locate the electron beam on individual phases. In order to minimize volatile loss, a defocused beam was used to analyze feldspar and leucite, and the beam was rastered over an area of approximately $25 \mu\text{m}^2$ to analyze glasses.

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TABLE 1. Compositions of gel starting materials (wt%)

Bulk composition		SiO_2	Al_2O_3	CaO	K_2O	Q	An	Ks
H1	Theor	60.8	21.1	2.02	16.1	36.0	10.0	54.0
H2	Theor	55.3	24.0	2.02	18.8	27.0	10.0	63.0
	Meas	56.8	23.0	2.13	17.9	28.8	10.6	60.6
H3	Theor	52.5	25.4	2.02	20.1	22.5	10.0	67.5
H4	Theor	49.7	26.9	2.02	21.4	18.0	10.0	72.0
	Meas	50.9	26.0	2.14	20.7	19.1	10.7	70.2
H5	Theor	44.1	29.8	2.02	24.1	9.0	10.0	81.0

Note: All compositions are normalized to 100%. Theor = theoretical compositions of gels; Meas = measured compositions obtained by microprobe analysis of glasses prepared from gels.

Starting materials for the experiments were gels, prepared according to the method described by Hamilton and Henderson (1968). The theoretical compositions of the starting materials are given in Table 1. All compositions quoted throughout this paper are given in weight percent proportions. Also given in Table 1 are microprobe analyses of glasses prepared from the gel starting materials for bulk compositions H2 and H4. Glasses were obtained in experiments which were above the liquidus temperature of the bulk compositions, at 795 °C and $P(\text{H}_2\text{O}) = 5$ kbar. The small, but consistent, discrepancies between the theoretical and analyzed compositions correspond to a loss of the kalsilite component in the analyzed compositions. This can also be seen in Figure 1. The losses of K and Al probably occur during gel preparation, and the analyzed glass compositions are thought to be more accurate as bulk compositions (Jones and MacKenzie, 1989). These small deviations from the theoretical composition do not represent a significant departure of the bulk composition from the plane Q-An-Ks: they only move the bulk compositions slightly away from the Ks corner of the ternary system relative to the theoretical composition.

Experiments of 4 d were found to give reversible results for gel, glass, and crystalline starting materials in the quaternary system Q-An-Ne-Ks studied by Jones and MacKenzie (1989), and were also considered to be of sufficient length for equilibrium to be achieved in the ternary system Q-An-Ks in this study. Microprobe analyses of products were stoichiometric. Euhedral crystals of all crystalline phases were observed, and no zoning was apparent. Kalsilite showed only limited solid solution with both anorthite and silica; leucite showed little deviation from the stoichiometric formula, KAlSi_2O_6 . When an assemblage of two feldspars + liquid was obtained, the amount of solid solution with the minor component (Or in plagioclase, An in alkali feldspar) was higher than that expected from previous studies of the limits of ternary solid solution in the feldspar system (Yoder et al., 1957; Seck, 1971). For example, in H1 at 785 °C plagioclase contains 19 wt% Or (Table 2). Two-feldspar pairs in this system have been shown to crystallize initially as fine intergrowths (Jones, 1989), and the origin of the apparent

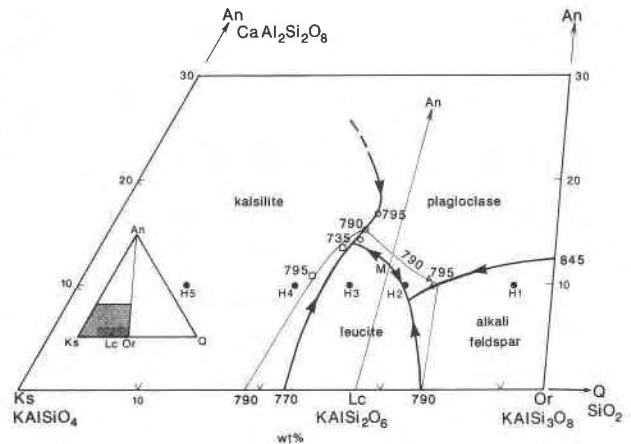


Fig. 1. Phase relationships in the system KAlSi_3O_8 - $\text{CaAl}_2\text{Si}_2\text{O}_8$ - KAlSiO_4 (Or-An-Ks) at $P(\text{H}_2\text{O}) = 5$ kbar. Filled circles = theoretical bulk compositions of starting materials (see Table 1). Open symbols = microprobe analyses of liquid compositions obtained from H2 (triangles), H4 (squares), and H5 (circles) at the temperatures shown (°C); see Table 2. Arrowed curves = inferred field boundaries. Also shown is the position of the 790 °C liquidus isotherm. The tie line joins leucite to anorthite, intersecting the plagioclase-leucite field boundary at the approximate position of the temperature maximum (M). Compositions H2 and H4 at 795 °C lie above the liquidus, and microprobe analyses of liquids show that the true bulk compositions are slightly kalsilite deficient compared with the theoretical compositions. Data on the joins $\text{CaAl}_2\text{Si}_2\text{O}_8$ - KAlSi_3O_8 (An-Or) and KAlSi_3O_8 - KAlSiO_4 (Or-Ks) are taken from Yoder et al. (1957) and Zeng and MacKenzie (1984) respectively.

excess of the third component is probably attributable to the presence of small quantities of intergrowths of the two feldspars in each feldspar phase.

RESULTS

Results obtained for bulk compositions H1-H5 are summarized in Table 2. Compositions of liquid, leucite, and kalsilite determined by microprobe analysis are recalculated in terms of the three components Q, An, and Ks so that they may be plotted in the ternary system. Feldspar analyses are given in terms of the feldspar components An and Or. When only a small quantity of glass was present, it could be observed optically but was difficult to analyze by microprobe because no areas of glass were sufficiently large. Liquid compositions determined by microprobe analysis usually gave totals of around 90 wt%; the shortfall reflects dissolved H_2O component.

The temperature for one experiment for bulk composition H2 is poorly constrained because there was a temperature gradient in the sample holder during the experiment. A minimum temperature of 735 °C was measured with one thermocouple. The other two results for H2 given in Table 2 indicate that the temperature for this experiment must have been between 735 and 795 °C. This data point has been included because it constrains the position of the two-feldspar field boundary in the ternary system (Fig. 1).

TABLE 2. Liquidus data in the system Q-An-Ks

Bulk composition	T (°C)	Phase	n	SiO ₂	Al ₂ O ₃	CaO	K ₂ O	Total	Q	An An	Ks Or	
H1	795	liq										
		plag	3	43.9	35.0	18.9	0.95	98.75		94.4	5.6	
			a.f.	4	64.0	17.9	0.44	14.6	96.94	2.5	97.5	
	785	liq										
		plag	4	47.4	32.8	16.4	3.19	99.79		81.2	18.8	
			a.f.	9	63.7	19.9	0.62	15.7	99.92	3.3	96.7	
735	leuc											
	plag											
	a.f.											
H2	795	liq	11	50.2	20.3	1.88	15.8	88.18	28.8	10.6	60.6	
		liq	7	52.6	21.3	1.83	16.6	92.33	29.3	9.9	60.8	
			plag	3	44.6	36.1	19.6	0.93	101.23		94.7	5.3
			a.f.	7	65.9	18.8	0.22	15.8	100.72		1.1	98.9
	735	leuc										
		plag										
a.f.												
H3	735	liq										
		leuc										
		plag										
H4	795	liq	10	44.8	22.9	1.88	18.2	87.78	19.1	10.7	70.2	
	735	liq	10	45.5	23.5	2.17	18.0	89.17	20.2	13.5	66.3	
		leuc	5	54.5	23.4	0.19	21.1	99.19	27.3	0.9	71.8	
		kals	3	38.7	31.9	0.13	29.2	99.93	0.7	0.7	98.6	
		plag	2	43.0	36.0	19.5	0.55	99.05		96.7	3.3	
H5	795	liq	10	45.9	23.7	3.01	16.2	88.81	21.4	16.9	61.7	
		kals	4	38.1	31.8	0.15	28.5	98.55	2.0	0.7	97.3	
		plag	5	44.0	35.9	19.1	1.06	100.06		93.8	6.2	
	790	liq	8	46.9	24.2	2.82	17.2	91.12	21.0	15.4	63.6	
		kals	5	38.7	32.0	0.11	29.0	99.81	1.7	0.5	97.8	
	735	plag	3	44.5	35.8	18.7	1.48	100.48		91.4	8.6	
		liq	10	46.8	23.4	2.56	17.0	89.76	21.3	14.3	64.4	
		leuc	2	54.2	23.2	0.25	21.0	98.65	26.9	1.3	71.8	
		kals	6	38.9	31.6	0.20	28.9	99.60	1.1	0.9	98.0	
		plag	5	44.1	36.0	19.6	0.95	100.65		94.5	5.5	

Note: $P(\text{H}_2\text{O}) = 5$ kbar. All data are in weight percent. Liquid identified as glass by optical examination only is indicated. XRD = phases identified by X-ray diffraction. Abbreviations: liq = liquid, plag = plagioclase, a.f. = alkali feldspar, leuc = leucite, kals = kalsilite, n = no. of analyses. Feldspars recalculated to normative feldspar components An-Or; all other phases recalculated to normative Q-An-Ks.

* Temperature lies between 735 and 795 °C (see text).

In Figure 1 the data of Table 2 are plotted in the ternary system Or-An-Ks. Liquidus data along the An-Or and Or-Ks joins are taken from Yoder et al. (1957) and Zeng and MacKenzie (1984) respectively. The positions of the two-feldspar and plagioclase + kalsilite field boundaries, and the position of the 790 °C isotherm in the kalsilite field are constrained by data in Table 2. Other liquidus field boundaries and the remainder of the 790 °C isotherm have been deduced to correlate with the data in Table 2. The ternary eutectic for plagioclase + leucite + kalsilite is defined by both compositions H4 and H5 at 735 °C. The liquid compositions obtained in these two products are in good agreement, and the composition of this eutectic shown in Figure 1 is the mean of the liquid compositions obtained from the two.

The leucite-plagioclase tie line of a leucite + plagioclase + liquid assemblage, such as that obtained from bulk composition H3 at 735 °C, intersects the plagioclase-leucite field boundary, as shown. The part of the tie line drawn in Figure 1 joins stoichiometric leucite with pure anorthite. Plagioclase compositions obtained in this study generally contain less than 10 wt% Or (Table 2), and the variation in the plagioclase composition between 90 and 100% An moves the position of the intersection of the

tie line with the plagioclase-leucite field boundary only slightly. This intersection necessitates a temperature maximum on the plagioclase-leucite field boundary close to the position indicated in Figure 1 as "M." Ternary eutectics occur on either side of this temperature maximum, viz., alkali feldspar + plagioclase + leucite + liquid and plagioclase + kalsilite + leucite + liquid. Both of these assemblages were obtained from the compositions studied. The former eutectic has a composition of approximately $\text{Q}_{28}\text{An}_8\text{Ks}_{64}$, read from Figure 1, and lies at a temperature slightly higher than 735 °C. The latter occurs at approximately $\text{Q}_{21}\text{An}_{14}\text{Ks}_{65}$ and is defined by bulk compositions H4 and H5 at 735 °C, which both contain the eutectic assemblage.

Bulk composition H3 would be expected to contain kalsilite at 735 °C, which is the temperature of the plagioclase + leucite + kalsilite eutectic, but none has been detected in the experimental products (Table 2). The apparent absence of kalsilite in this sample can probably be accounted for by the fact that the true bulk compositions are depleted in the kalsilite component relative to theoretical bulk compositions, as described above. If composition H3 is depleted in Ks in proportions similar to those of H2 and H4 (Table 1 and Fig. 1), its true com-

TABLE 3. Normative felsic components of Roccamonfina lavas, after Appleton (1972)

Rock type	Or	Ab	An	Lc	Ne	Total
LCT	12.2	0.0	16.4	21.5	7.5	57.6
TL	33.9	0.0	16.1	16.3	11.2	77.5
PLT	51.5	6.6	14.1	0.0	12.2	84.4
TLP	55.0	16.8	14.2	0.0	5.9	91.9
LP	56.4	19.2	9.8	0.0	11.4	96.8

Note: LCT = leucitite; TL = tephritic leucitite; PLT = phonolitic leucite tephrite; TLP = tephritic leucite phonolite; LP = leucite phonolite.

position probably lies close to the anorthite-leucite tie line and only minor proportions of kalsilite would be expected to be present at 735 °C.

Liquidus temperatures and the positions of phase boundaries determined in this study are consistent with previously determined phase relationships along the binary joins An-Or (Yoder et al., 1957) and Or-Ks (Zeng and MacKenzie, 1984) and also correspond with phase relationships in the quaternary system Q-An-Ne-Ks at $P(\text{H}_2\text{O}) = 5$ kbar described by Jones and MacKenzie (1989).

PETROLOGICAL APPLICATION

The ternary system Q-An-Ks has been used previously in descriptions of the crystallization behavior of silica-undersaturated, K-rich rocks. Appleton (1972) discussed the fractionation trend observed in the high-K series of the Roccamonfina volcanic complex, Italy, in terms of this system. The fractionation series of leucitite (LCT)-tephritic leucitite (TL)-phonolitic leucite tephrite (PLT)-tephritic leucite phonolite (TLP)-leucite phonolite (LP) was described by Appleton (1972) as representing fractionation of biotite-pyroxenite followed by biotite-gabbro at a pressure sufficiently high to prevent the crystallization of leucite.

We have determined average compositions of the above rock types in the Roccamonfina complex from data for individual analyses given by Appleton (1972). Normative felsic components (An + Ab + Or + Lc + Ne) of these rocks are given in Table 3. None of these rocks contain normative Qz or Ks. The sum of the felsic components is 58 wt% for leucitite, and >75% for the other rock types. The components An, Or, and Lc, represented by the system Q-An-Lc, total >65 wt% of the whole rock for the TL, PLT, TLP, and LP. Thus, application of phase relationships in this ternary system may be expected to be appropriate in determining fractionation trends in the Roccamonfina lavas.

The 5 kbar phase relationships we have determined in this study are H_2O saturated. This condition may not be directly applicable during fractionation of the Roccamonfina suite, although the presence of biotite and phlogopite phenocrysts is evidence that a significant H_2O component was present. Even though liquidus temperatures and the absolute positions of the phase boundaries may not be known exactly for H_2O -undersaturated conditions, the

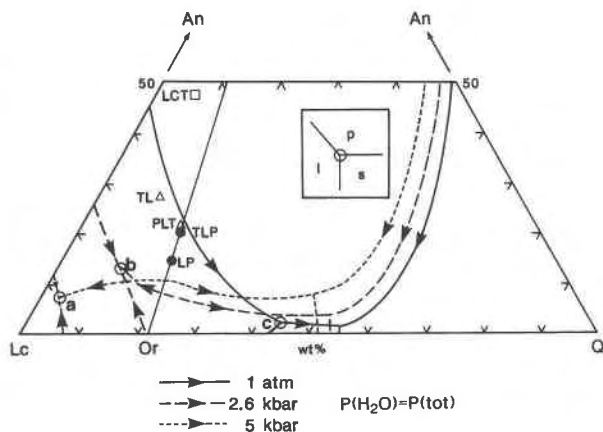


Fig. 2. Roccamonfina high-K lavas plotted in terms of the system Q-An-Lc. Field boundaries at $P(\text{H}_2\text{O}) = 1$ bar (solid lines) are from Schairer and Bowen (1947); those at 2.6 kbar (long-dashed lines) are based on boundaries deduced by Appleton (1972) but are revised to show temperatures decreasing from the two-feldspar plane in both directions. Field boundaries at 5 kbar (short-dashed lines) are from Figure 1. Extensions of 2.6 and 5 kbar boundaries into the silica-oversaturated part of the system (Q-An-Or) are schematic only. Mean compositions of the following rock types are plotted: leucitite (LCT), tephritic leucitite (TL), phonolitic leucite tephrite (PLT), tephritic leucite phonolite (TLP), leucite phonolite (LP) (see Table 3). Sums of normative felsic components are: square <75%, triangles 75–90%, filled circles >90%. Open circles labeled a, b, c, are at points where fields of plagioclase (p), leucite (l), and sanidine (s) coexist in the topological relationship shown schematically in the inset.

general form of phase relationships has been shown not to change significantly within this system for $P(\text{H}_2\text{O}) < P(\text{total})$. Zeng (1983) showed that, for $P(\text{total}) = 5$ kbar in the system Q-Ne-Ks when $P(\text{H}_2\text{O}) < P(\text{total})$, the stability field of leucite expands and liquidus temperatures are higher relative to the H_2O -saturated system. Thus, the form of phase relationships at a lower pressure of H_2O in the ternary system Q-An-Ks at $P(\text{total}) = 5$ kbar is likely to be approximately equivalent to H_2O -saturated phase relationships at lower total pressure, e.g., 2.6 kbar (Fig. 2; Appleton, 1972). Because other volatiles may also play a role in modifying phase relationships, we discuss the system here in terms of $P(\text{H}_2\text{O}) = P(\text{total})$. Although the absolute total pressure may vary, the form of the phase relationships is the most important factor in determining fractionation trends.

Appleton (1972) discussed the fractionation trend in the high-K series in terms of the ternary system Q-An-Lc at $P(\text{H}_2\text{O}) = 2.6$ kbar. The phase relationships that Appleton deduced for this pressure (Fig. 2) are incorrect, however. He showed temperatures on the plagioclase-sanidine field boundary falling along its entire length. This is not to be expected from experimental studies that have shown that within the stability field of feldspar in this system, the ternary feldspar plane acts as a thermal bar-

rier, and the temperatures decrease from the ternary feldspar plane in both directions (Morse, 1968; Norris and MacKenzie, 1976). The revised form of the field boundaries at 2.6 kbar is shown in Figure 2, along with field boundaries determined in this study at 5 kbar. These latter boundaries have been extended schematically into the silica-oversaturated portion of the system, where the expansion of the quartz field relative to that of feldspar with increasing pressure is expected from known phase relationships in the granite system (Tuttle and Bowen, 1958). According to Appleton's version of the diagram at 2.6 kbar, a bulk composition plotting in the plagioclase field, such as TL, will crystallize plagioclase followed by either leucite or sanidine. Under fractionating conditions, liquids would be expected to progress along the two-feldspar boundary, with no barrier to the formation of silica-oversaturated liquids. However, no evidence for silica-oversaturated liquids was reported by Appleton (1972) in the high-K series of the Roccamonfina lavas. Phase relationships are, therefore, more likely to be of the form determined at $P(\text{H}_2\text{O}) = 5$ kbar in this study or those shown for 2.6 kbar in Figure 2. These relationships are also consistent with phase relationships under H_2O -undersaturated conditions at higher total pressure as discussed above. A liquid plotting in the plagioclase field, such as TL, will crystallize plagioclase followed by sanidine then leucite, or vice versa, and remain silica undersaturated. Only at lower pressures are magmas with compositions similar to those of the high-K Roccamonfina lavas likely to produce silica-saturated differentiates.

Phase relationships in the ternary system Q-An-Lc are valuable to the interpretation of broad fractionation trends in the Roccamonfina lavas. However, some details may be obscured because all the rock types have a small but significant nepheline component in their normative compositions (Table 3). Jones and MacKenzie (1989) considered the same rock compositions in terms of the quaternary system Q-An-Ne-Ks and showed that the derivation of leucite phonolite from tephritic leucite phonolite may be controlled by the fractionation of two feldspars at pressures high enough to suppress the stability field of leucite. This relationship is not apparent in the ternary system Q-An-Lc.

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