

Leucite–Na-feldspar incompatibility: an experimental study

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SUMMARY. Phase relations at atmospheric pressure in the pseudobinary join KAlSi_3O_8 (Lc)– $\text{NaAlSi}_3\text{O}_8$ (Ab) and in the pseudoternary join Lc–Ab– $\text{CaAl}_2\text{Si}_2\text{O}_8$ (An) indicate that leucite is incompatible with Na-feldspar. In the former join leucite can exist with an alkali feldspar of maximum albite content Ab_{54} . In the Lc–Ab–An join, leucite only coexists with ternary feldspars with high An contents (approximately An_{50}). Under $P_{\text{H}_2\text{O}}$ conditions leucites may only coexist with alkali feldspars even poorer in Ab than those found at atmospheric pressure. Rare occurrences of coexisting leucite and Na-feldspar in nature have probably not crystallized directly from a melt but may have formed by a process of alkali ion exchange; or they may be unstable assemblages. No support can be found for the suggestions based on thermochemical calculations that albite and leucite are compatible at high temperatures.

In leucite-bearing lavas the coexisting feldspar is either a fairly Ca-rich plagioclase or a K-rich alkali feldspar or both (Shand, 1947). On thermochemical grounds Miyashiro (1960) has postulated that the apparent incompatibility between leucite and albite may not exist at high temperatures. MacKenzie and Rahman (1968) describe veins of leucite and Na-feldspar in a basanite from the Massif Central, France, but were unable to confirm this as a stable assemblage. W. S. Fyfe (quote in MacKenzie and Rahman (1968)) calculated that the leucite–albite assemblage may become stable around 1000 °C. Experimental studies at $P_{\text{H}_2\text{O}}$ up to 1000 bars in the K-rich portion of the NaAlSiO_4 – KAlSiO_4 – SiO_2 – H_2O system (Fudali, 1963) indicate low melting temperatures for bulk compositions likely to contain the leucite–albite pair, and at lower water pressures the stable assemblage for bulk compositions likely to crystallize leucite and albite to be nepheline, a K-rich feldspar, and a Na-rich leucite. The only other studies in this system are liquidus relations at atmospheric pressure (Schaerer, 1957).

In an attempt to elucidate the leucite–Na-feldspar incompatibility and to determine the limits of plagioclase and alkali feldspar compositions that may coexist with leucite in volcanic rocks, a study of phase relations in the systems Lc–Ab and Lc–Ab–An has been made at 1 atmosphere pressure. The former represents a pseudobinary join in the Na_2O – K_2O – Al_2O_3 – SiO_2 system in which subliquidus and solidus temperatures are sufficiently high to resolve the suggestion (Miyashiro, 1960; Fyfe (in MacKenzie and Rahman, 1968)) that leucite and Na-feldspar may coexist at high temperatures.

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The Lc–Ab–An system is a pseudoternary join in the system $\text{CaO–Al}_2\text{O}_3\text{–K}_2\text{O–Na}_2\text{O–SiO}_2$; the bounding joins Lc–An and Ab–An in this system have been determined by Schairer and Bowen (1947) and Bowen (1913) respectively.

Experimental methods

Preparation of starting materials and experimental techniques. The high viscosity of liquids encountered in alkali-rich compositions in these joins makes determination of equilibrium difficult. Since complete crystallization of these liquids at atmospheric pressure is extremely slow (Schairer, 1950), the starting materials, prepared as homogeneous glasses (Schairer, 1959) or as sintered materials (near the Lc-rich end of the join Lc–Ab), were completely crystallized in sealed gold tubes at 700 °C under 1 Kb $P_{\text{H}_2\text{O}}$ for 3 to 14 days before use. With such starting materials equilibrium is more likely to be attained. The possibility that alkalis and other components may have been lost to a vapour phase during hydrothermal crystallization cannot be ignored but if present is believed to be minimal. Absence of corroded crystals, agreement of data with that of nearby mixtures, good distribution of the phases in glass, and reversibility of reactions across boundaries suggest that equilibrium was obtained.

All quenching experiments were done in voltage stabilized vertical resistance furnaces. Temperatures were measured by frequently calibrated Pt–Pt₉₀Rh₁₀ thermocouples and are believed accurate to within ± 5 °C.

Crystalline phases in these systems are feldspars, nepheline, leucite, and orthorhombic kalsilite, all of which are complex solid solutions. Identification was made by optical or X-ray diffraction methods or both.

Determination of feldspar and nepheline compositions. The principal objective of this study was to determine the compositions of feldspars coexisting with leucite. In the Lc–Ab–An join, the feldspars are ternary and amenable to microprobe analyses. However in the Lc–Ab join the alkali feldspars could not be accurately analysed by microprobe due to alkali volatilization. Variations in the compositions of feldspars in this join with varying bulk composition were determined by measurements of cell constants (Orville, 1967). Samples were exposed to Fe- $K\alpha$ radiation in a Guinier camera for 20 to 45 hours using an internal silicon standard. The position of each line of the feldspar was determined by a least squares best fit computer method comparing measured lines and known lines of pure silicon. A further programme calculated the lattice constants and their probable errors by a least squares method; from these, cell volumes of the alkali feldspars were determined.

Selected nepheline compositions were determined by measurement of the 20 $\bar{2}2$ and 21 $\bar{3}0$ reflections using a Guinier camera and the curves of Hamilton and MacKenzie (1960).

Other crystalline phases. Other phases are leucite and orthorhombic kalsilite solid solutions. Leucite is easily distinguished optically from kalsilite and nepheline by its near isotropism, whereas nepheline could only be distinguished from kalsilite by the 222 and 421 reflections of kalsilite (Smith and Tuttle, 1957). The extent of leucite solid solution is much less in this system than in the leucite–analcime system (Fudali, 1963) as shown by increases in the 211, 004, 400, and 420 spacings.

Experimental results

The join Lc-Ab. The data in Table I and fig. 1 indicate that this join is a pseudobinary join cutting the primary phase volumes of feldspar and leucite in the quaternary system $\text{Na}_2\text{O}-\text{K}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$. Point A ($\text{Lc}_{41}\text{Ab}_{59}$, 1068 ± 5 °C) represents a pseudoeutectic where leucite and feldspar coexist with liquid. The temperature is in good agreement with the extrapolated value obtained from the same composition in the $\text{NaAl-SiO}_4\text{-KAlSiO}_4\text{-SiO}_2$ system (Schairer, 1957, fig. 29). Point B, at the same composition

TABLE I. *Quenching data* on the system Lc-Ab-An*

Refractive index†	Composition (wt.%)			Liquidus temp.	Primary phase	Other phases‡
	Lc	Ab	An			
1.490	20	80	—	1110 °C	Felds	Ne in 1015. B.M. 1000
1.487	30	70	—	1100	Felds	Ne in 1010. B.M. 985
1.488	40	60	—	1075	Felds	Ne in 1040, Lc in 995, B.M. 990
1.489	45	55	—	1220	Lc	Ne in 1152, Felds in 1065
1.487	50	50	—	1280	Lc	Ne in 1190, Felds in 1090, B.M. 1015
1.490	60	40	—	1385	Lc	Ne in 1270, Felds in 1095. B.M. 1035.
—	70	30	—	nd	Lc	At 1380, Lc. At 1145 Lc-Ne. At 1050, Lc-Ne-Feld.
—	80	20	—	nd	Lc	Ne in 1430, Felds in 1105. B.M. 1085.
—	85	15	—	nd	Lc	OKs in 1475, Felds in 1085, Lc-OKs-Felds at 1075.
—	90	10	—	nd	Lc	At 1480, Lc-OKs-L, Felds in 1030, at 1015 Lc-OKs-Felds
—	95	5	—	nd	Lc	At 1480 Lc-OKs-L. Felds in 975.
1.491	47	16	37	1374	Felds	Lc in 1370
1.505	43	31	26	1330	Lc	Felds in 1326
1.501	40	44	16	1285	Felds	Lc in 1270
1.495	40	50	10	1210	Felds	Lc in 1205
1.496	42	53	5	1190	Felds	Lc in 1180
1.497	30	60	10	1220	Felds	

* Runs producing only liquid are not given but are included in fig. 1. Times of runs varied from 1 to 5 hours to minimize alkali loss (see Gupta, 1972).

† Refractive index of glass prior to crystallization. Compositions with no R.I.'s are sintered materials.

‡ Abbreviations: Felds—feldspar, Ne—nepheline, Lc—leucite, OKs—orthorhombic kalsilite, B.M.—beginning of melting.

and 1030 ± 5 °C, is a piercing point representing the intersection of the Lc-Ab system with a univariant line in the $\text{Na}_2\text{O}-\text{K}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$ system where leucite, nepheline, and feldspar coexist with liquid. Point C (990 ± 5 °C) is the intersection of the solidus in the system and the composition at which leucite and feldspar are no longer mutually stable phases.

As shown in fig. 1, the extent of solid solution of albite in leucite is very small and cannot extend beyond $\text{Lc}_{95}\text{Ab}_5$ at temperatures greater than 1020 °C, where leucite and orthorhombic kalsilite solid solutions exist. This minor solid solution is in contrast to the large leucite solid solution field in the leucite-analcime join at 1 kb $P_{\text{H}_2\text{O}}$

found by Fudali (1963), who suggested that extensive solid solution was restricted to this join (Fudali, 1963, p. 1105). A slight increase in the interplaner spacings of the 211, 004, 400, and 420 of leucites crystallized from bulk compositions $Lc_{95}Ab_5$ and $Lc_{90}Ab_{10}$ compared with those for pure leucite, and a decrease in the 132 and 202

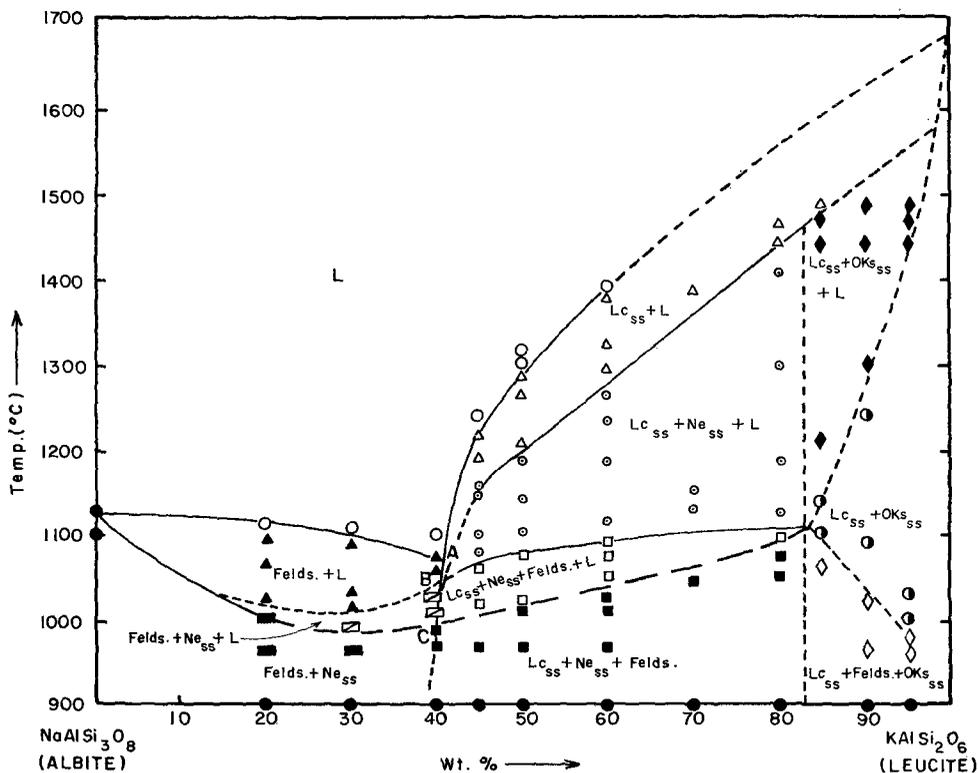


FIG. 1. Phase relations in the system Lc-Ab. A, B, and C are discussed in text. Abbreviations are given in Table I. Dashed lines are extrapolations.

spacings of kalsilite from the same compositions relative to pure kalsilite (Smith and Tuttle, 1957) indicates that both these minerals are solid solutions. The nature of these solid solutions could not be determined.

Compositions of alkali feldspars in the Lc-Ab join. In this join feldspars may coexist with leucite in the absence of liquid between $Ab_{59}Lc_{41}$ to Ab_5Lc_{95} below about 1000 to 1075 °C depending on the bulk composition (fig. 1). Variations in cell parameters of six feldspars crystallized at 960 °C from Ab_{100} to $Ab_{10}Lc_{90}$ are shown in fig. 2a-f with corresponding cell volumes and compositions determined by the method of Orville (1967, fig. 5), given in Table II and fig. 2g. Fig. 2d and f indicate a discontinuity in the α and γ angles of the feldspars at about $Ab_{60}Lc_{40}$ indicating their change from triclinic to monoclinic symmetry.

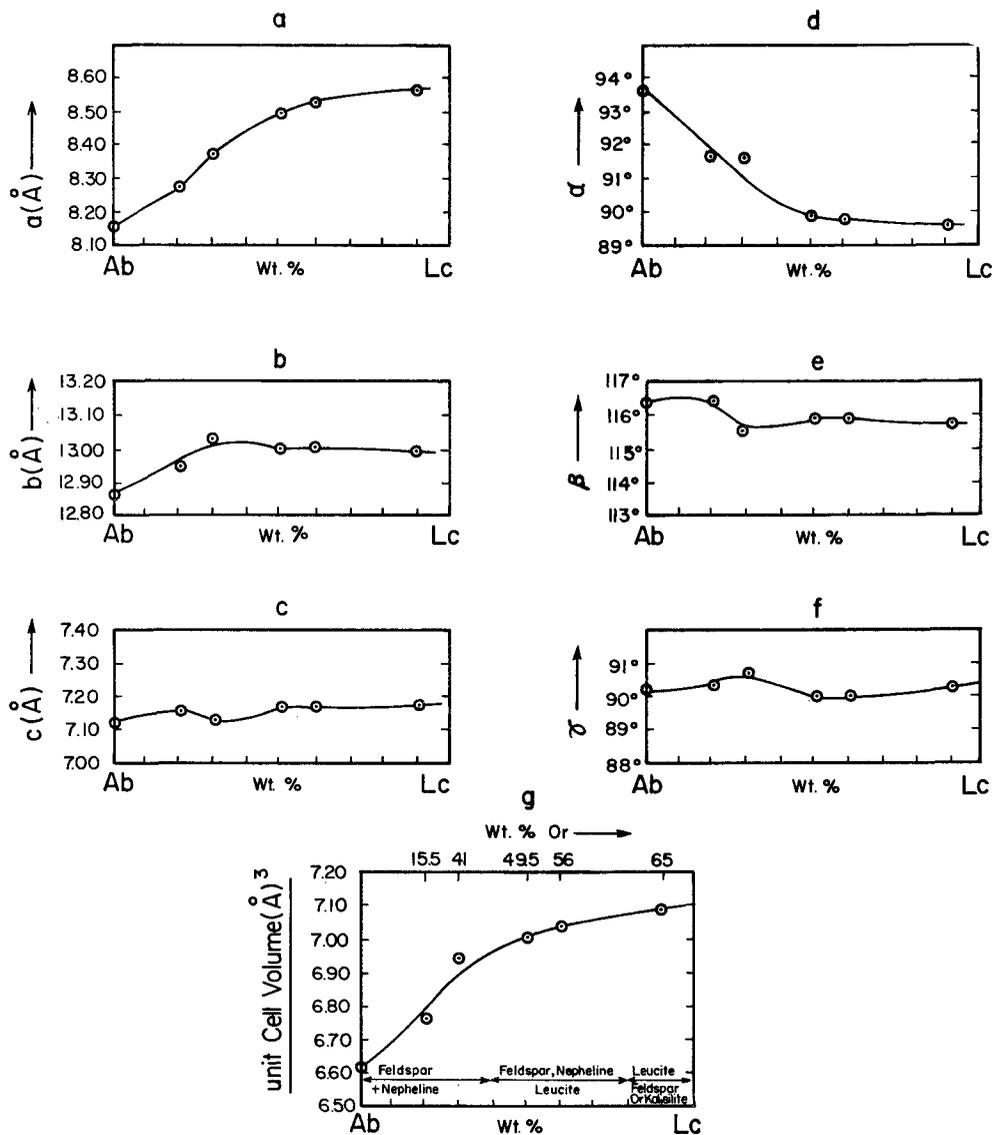


FIG. 2a-g. Cell constants of feldspars crystallized in the system Lc-Ab at 960 °C.

Extrapolation of the unit cell volume of the feldspar crystallizing from the composition $Ab_{59}Lc_{41}$ (fig. 2g) gives a corresponding feldspar of composition $Ab_{54}Or_{46}$ and represents the most Na-rich feldspar that may crystallize with leucite at subsolidus temperatures in this join. With increasing K_2O in bulk compositions and concomitant crystallization of leucite and eventually orthorhombic kalsilite, the rate of increase in the Or component of the crystallizing feldspar decreases (figs. 1 and 2g).

TABLE II. Compositions of feldspars in the join Lc–Ab
Crystallized at 1 atm and 960 °C

Bulk composition		Cell vol.	Composition as wt.%, determined from cell volume, after Orville, 1967	
Ab	Lc		Ab	Or
100	0	661.51 Å ³	100	—
80	20	677.19	84.5	15.5
70	30	694.49	59	41
50	50	700.19	50.5	49.5
40	60	704.26	44	56
10	90	709.05	35	65

The composition of nepheline coexisting with feldspar only at 960 °C from the composition Ab₇₀Lc₃₀ gave Ne₆₈Ks₃₀Qz₂. When this composition is plotted along with the coexisting feldspar composition (Ab₅₉Or₄₁—Table II) on the system NaAlSi₃O₈–KAlSi₂O₆–SiO₂ the tie line passes very close to the bulk composition Ab₇₀Lc₃₀

indicating the absence of any other phase. Compositions of nephelines crystallized at the same temperature from bulk compositions Ab₆₀Lc₄₀ and Ab₅₀Lc₅₀ (where leucite is an additional phase) gave the same value (Ne₇₃Ks₂₅Qz₃).

The join Lc–Ab–An. Liquidus relations in this pseudoternary portion of the system CaO–Al₂O₃–Na₂O–K₂O–SiO₂ are given in Table I and fig. 3. Due to the pseudoternary nature of this join liquids cannot be represented by the Lc–Ab–An compositional plane. Estimations of the nature of these liquids can be made from determinations of the compositions of feldspars crystallized at or near the line A–B (fig. 3).

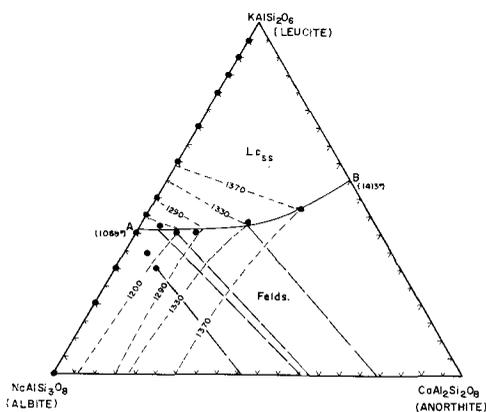


FIG. 3. Liquidus relations in the system Lc–Ab–An. Heavy dashed lines indicate compositions of ternary feldspars (projected on to Ab–An join) crystallized at various temperatures (see Table III).

Compositions of ternary feldspars in the Lc–Ab–An join. Microprobe analyses of feldspars were determined from four bulk compositions crystallized at temperatures close to liquidus ranging from 1320 to 1180 °C (Table III). In fig. 3 tie lines have been drawn between these bulk compositions and the corresponding feldspar composition (as plagioclase only) projected on to the Lc–Ab–An join. These tie lines show progressive enrichment in the (Ab+Or) content of the feldspar with decreasing temperature but indicate that the plagioclase coexisting with a liquid low in An content (Lc₅₃Ab₄₂An₅) is rich in An (An₆₁, Ab+Or₃₉). This implies that the residual liquid after crystallization of such a feldspar must be

enriched in $\text{NaAlSi}_3\text{O}_8$ although small amounts of soda may be incorporated in leucite. These results show that the addition of anorthite does not promote leucite-Na-feldspar compatibility.

TABLE III. *Microprobe analyses* of feldspars in the system Lc-Ab-An*

No.	Composition of mixtures			Temp.	Wt. %				An wt %	Ab+Or wt %
	Lc	Ab	An		SiO ₂	Na ₂ O+K ₂ O	Al ₂ O ₃	CaO		
1	43	31	26	1320 °C	45.92	4.54	33.32	16.22	80.40	19.60
2	40	50	10	1180	51.87	5.72	29.64	12.77	63.15	36.85
3	53	42	5	1180	50.35	7.05	30.29	12.31	60.93	39.07
4	30	60	10	1200	57.09	5.44	28.23	9.24	45.63	54.37

* CaO, Al₂O₃, and SiO₂ determined; Na₂O+K₂O by difference.

Petrological implications

Study of phase relations in the joins Lc-Ab and Lc-Ab-An at atmospheric pressure support the conclusions of field studies that leucite and Na-rich feldspar are incompatible. In the absence of the An molecule the most Na-rich feldspar that can coexist with leucite contains 46 wt. % Or. In the presence of the An molecule, leucites coexist only with ternary feldspars containing high proportions of anorthitic plagioclase (approximately An₅₀, Ab+Or₅₀). The incompatibility of leucite and albitic feldspar in the absence of the An molecule is probably due to a reaction of the type:



Thus residual liquids produced by such a reaction will be enriched in silica, although some of the silica may be incorporated by nepheline. In the Lc-Ab-An system, more closely representing more basic liquids from which leucite-feldspar assemblages crystallize, the products of crystallization at low temperatures near the liquidus surface are an An-rich ternary feldspar, leucite, and a residual liquid enriched in Na₂O and SiO₂. Some of this Na₂O may be incorporated into leucite. MacKenzie and Rahman (1968) noted that the leucite rims in the leucite-Na-feldspar veins in the Massif Central basanite became enriched in Na₂O with falling temperature. The presence of a Na₂O-enriched residual liquid might also explain the albitic nature of the feldspar in these veins, the Na-rich feldspar having formed from an original K-rich feldspar by a process of alkali ion exchange. Such a mechanism would explain the maximum Or content of 20 mol. % (MacKenzie and Rahman, 1968) relative to the Ab-Or content of 50 wt. % expected from the direct primary crystallization of low-temperature liquids in the simplified Lc-Ab-An join.

The effect of $P_{\text{H}_2\text{O}}$ on the incompatibility of leucite and Na-feldspar may be estimated by comparing the results of the present study of the Lc-Ab join at atmospheric pressure with those of Fudali (1963, Fig. 4A) for the 850 °C isothermal section of the

NaAlSiO₄-KAlSiO₄-SiO₂ at $P_{\text{H}_2\text{O}} = 265$ bar where the stable assemblage for a bulk composition of approximately Ab₅₀Lc₅₀ is leucite, nepheline, and feldspar of composition about Or₆₀Ab₄₀. Table II shows that the corresponding bulk composition at atmospheric pressure crystallizes leucite, nepheline, and feldspar of composition Or_{49.5}Ab_{50.5}. Thus crystallization under modest $P_{\text{H}_2\text{O}}$ conditions appears to decrease the chances of compatibility of leucite and Na-feldspar. Under higher $P_{\text{H}_2\text{O}}$ the stability of leucite decreases (Tuttle and Bowen, 1958) and crystallization of leucite-feldspar assemblages will be restricted to very K-rich bulk compositions.

The compositions of feldspars coexisting with leucites (Tables II and III) at temperatures in excess of 1000 °C does not support the suggestions of Miyashiro (1960) and Fyfe (in MacKenzie and Rahman, 1968), based on thermochemical considerations, that leucite and Na-feldspar are compatible at high temperatures.

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