# AN EXPERIMENTAL STUDY ON THE LEUCITE-PSEUDOLEUCITE PROBLEM[1]

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ABSTRACT

Experimental works on leucite using a piston-anvil device shows the following chemical reaction is favored by increasing pressure and/or decreasing temperature:

$$2 \text{ leucite} \rightarrow \text{sanidine} + \text{hexagonal kalsilite}$$
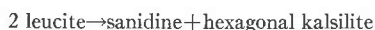
The genesis of pseudoleucite is discussed and it is concluded that the breakdown of leucite into a mixture of sanidine and hexagonal kalsilite-nepheline solid solution caused by falling temperature under a substantial partial pressure of water is one of the most important mechanisms by which pseudoleucite is formed in some igneous rocks.

INTRODUCTION

The formation of pseudoleucite, a mixture of potash-feldspar and nepheline-kalsilite solid solution, in potash-rich igneous rocks has been studied for the last half century by many authors. The explanations proposed by these authors can be divided into three groups:

(1) Unmixing breakdown of leucite into potash-feldspar and nepheline-kalsilite solid solution (Knight, 1906; Scharizer, 1928; Larsen and Buie, 1938; Larsen *et al.*, 1939, 1941; Yagi, 1954).

(2) Chemical reaction between leucite crystals and magmatic or hydrothermal residual solutions (Bowen and Ellestad, 1937; Schairer and Bowen, 1955; Zies and Chayes, 1960).

(3) Decomposition of leucite into potash-feldspar and nepheline-kalsilite solid solution (Larsen et al., 1938, 1939, 1941; Tilley, 1957; Fudali, 1957, 1963).

There is much petrographic evidence to indicate that pseudoleucite was usually formed after the magma reached its present position, after intrusion or extrusion, but before complete cooling of the rock. Hence the process of formation of pseudoleucite must have occurred within a cooling magma.

Though many geological, petrographical and mineralogical descriptions have been published, and many diverse opinions about the genesis of pseudoleucite have been presented, no experimental study over a wide range of temperature and pressure conditions has been published. In this paper we present the results of our experimental study on the stability of leucite over a temperature range of 200–850° C. and pressures up to 40 kilobars.

Experimental Methods and Starting Materials

Our experiments were performed using the "simple squeezer" piston-anvil device, which has been already described by Griggs and Kennedy (1956 and Pistorius *et al.* (1962). Samples to be subjected to high pressure in the "simple squeezer" were placed in platinum foils in the form of a wet powder. Water, acting as a flux, accelerates the reaction rate even though it does not participate in the reaction. In all runs, pressure was first raised to the desired value and then temperature was raised. The duration of our runs in the "simple squeezer" was 2 hours for temperatures above 500° C., 4 hours at 400–500° C. and 12–24 hours below 400° C.

Our starting material was a natural leucite obtained from the mineral collection of the Geology Department of the University of California, Los Angeles. Na_2O content of this leucite was less than 0.5%, *i.e.*, it has almost pure $KalSi_2O_5$ composition. X-ray powder data, unit-cell dimensions, and unit-cell volume of the leucite are shown in Table 1. These data also indicate that the leucite has almost pure leucite end member composition (Bannister, 1931; Prider and Cole, 1942).

Identification of materials formed in the experimental works was by x-ray diffractometer and the polarizing microscope. Quartz powder was used as an internal standard in detailed x-ray study.

Results

Results obtained from our experimental work are summarized in Fig. 1. Leucite has been broken down into an assemblage of sanidine and hexagonal kalsilite under relatively high pressures. The assemblage of sanidine and hexagonal kalsilite formed from leucite in the "simple squeezer" was also reacted and transformed into leucite at decreased pressure and/or with increasing temperature. Thus the boundary between the field of leucite and that of sanidine+hexagonal kalsilite presented in Fig. 1 is believed to be an equilibrium boundary.

Neither the breakdown of leucite into sanidine+hexagonal kalsilite nor the chemical reaction of sanidine and hexagonal kalsilite to form leucite occurred with dry starting materials even with experiments of 2 to 5 days duration and high temperatures and pressures. This suggests that water plays a very important role in accelerating the breakdown or the chemical reaction.

X-ray powder patterns, unit-cell dimensions and unit-cell volumes of sanidine and hexagonal kalsilite formed as breakdown products of leucite are shown in Tables 2 and 3. Judging from these data, the sanidine and kalsilite are almost pure $KalSi_3O_8$ and $KalSiO_4$ respectively, and are very low in soda (Sahama *et al.*, 1956; Smith and Sahama 1954; Smith and

TABLE 1. X-RAY POWDER DATA AND UNIT-CELL DIMENSIONS OF
LEUCITE USED AS A STARTING MATERIAL

hkl	Measured		Calculated	hkl	Measured		Calculated
	d(Å)	I	d(Å)		d(Å)	I	d(Å)
112	5.52	20	5.51	233 323	2.842	40	2.844
121 211	5.38	40	5.38	332	2.814	27	2.814
022 202	4.74	12	4.75	224	2.765	3	2.765
013 103	4.32	8	4.32	422 242 015 105	2.687	5	2.687
213 123	3.605	17	3.605	134 314	2.644	12	2.644
312 132	3.539	17	3.540	143 413	2.608	4	2.608
004	3.440	100	3.439	215 125	2.490	4	2.489
040 400	3.266	100	3.266	251 521	2.391	3	2.391
033 303	3.160	5	3.158	044 404	2.368	32	2.368
411 141	3.091	8	3.089	305 035	2.326	3	2.326
204 024	3.046	9	3.046	440	2.309	4	2.310
402 042	2.954	5	2.953	053 503 343 433	2.271	4	2.271
240 420	2.923	41	2.924				

$a = 13.07$ (0) Å
$c = 13.74$ (4) Å
Tetragonal
Unit-cell volume $= 146.7$ Å^3
$Na_2O < 0.5\%$ (wt)

TABLE 1—(*continued*)

hkl	Measured		Calculated	hkl	Measured		Calculated
	d(Å)	I	d(Å)		d(Å)	I	d(Å)
442 325 235	2.191	2	2.191	217 127	1.862	5	1.862
				336	1.839	4	1.839
026 206	2.162	5	2.162	640 460	1.814	17	1.813
253 523	2.144	3	2.145	037 307	1.791	3	1.791
523 352	2.130	5	2.132	552 172 712	1.785	4	1.785
620 260	2.067	4	2.067	271 721	1.780	4	1.781
514 154	2.055	4	2.055	237 327	1.727	8	1.727
136 316	2.005	2	2.004	372 732	1.665	18	1.665
107 017	1.943	3	1.942	651 561 028 208	1.662	15	1.662
444	1.916	4	1.917				
046 406	1.875	4	1.876	446	1.654	6	1.645

Tuttle, 1957), further proof of the low soda content of the starting leucite.

As shown in Fig. 1, leucite and the mixture of sanidine and hexagonal kalsilite made from leucite were transformed into the assemblage of hexagonal kalsilite and a new phase of the chemical composition of $KalSi_3O_8$ $\cdot H_2O$ under very high water pressures.

The x-ray data and other physical properties of the new phase, $KalSi_3O_8 \cdot H_2O$, made from $KalSi_3O_8$ glass $+ H_2O$ in the "simple squeezer", and the data for the hexagonal kalsilite in the breakdown products of leucite are shown in Table 4. The stability relations between the new phase and sanidine have been described in detail in another paper (Seki and Kennedy, 1964).

An assemblage of hexagonal kalsilite and $KAlSi_3O_8 \cdot H_2O$ formed from leucite was also easily and completely transformed into a mixture of hexagonal kalsilite and sanidine at water pressures lower than the upper pressure limit of sanidine. Thus, we believe that the boundary between the sanidine+hexagonal kalsilite field and $KAlSi_3O_8 \cdot H_2O$+hexagonal kalsil-
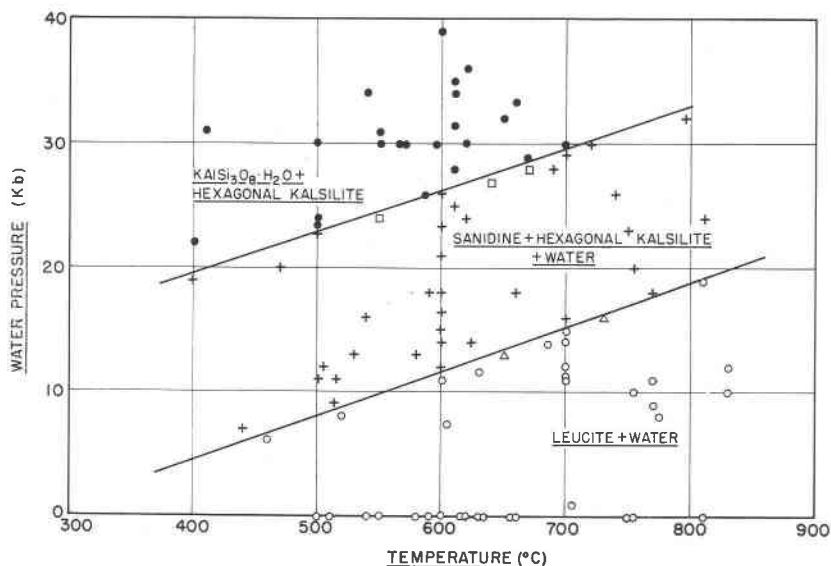

Fig. 1. Diagram showing the stability fields of leucite, sanidine+hexagonal kalsilite, and $KAlSi_3O_8 \cdot H_2O$+hexagonal kalsilite determined by means of the simple squeezer under wet condition.

○ leucite+water→leucite+water
△ sanidine+hexagonal kalsilite+water→leucite+water
+ leucite+water→sanidine+hexagonal kalsilite+water
□ $KAlSi_3O_8 \cdot H_2O$+hexagonal kalsilite→sanidine+hexagonal kalsilite+water
● leucite+water→$KAlSi_3O_8 \cdot H_2O$+hexagonal kalsilite

ite field in Fig. 1 is an equilibrium boundary for the chemical reaction sanidine+$H_2O \rightleftarrows KAlSi_3O_8 \cdot H_2O$.

Schairer and Bowen (1955) found in their experimental study of the system K_2O-Al_2O_3-SiO_2 that sanidine and hexagonal kalsilite are incompatible at high temperature (700–1700° C.) and atmospheric pressure. It is clear from this study, however, that these phases can coexist at high pressure.

DISCUSSION

Knight (1906) reported that leucite can have a moderate content of Na replacing K. The formation of pseudoleucite has often been thought as

TABLE 2. X-RAY POWDER DATA, UNIT-CELL DIMENSIONS
AND UNIT-CELL VOLUME OF SANIDINES

hkl	Sanidine synthesized from $KAlSi_3O_8$ glass (20 kb, 600° C.) (this paper)			Sanidine formed by breakdown of leucite (L.97, 29 kb, 700° C.) (this paper)	
	Measured		Calculated	d(Å)	I
	e(Å)	I	d(Å)		
110 $\overline{1}$10	6.68	7	6.68	6.63	9
020	6.51	4	6.50	6.51	9
$\overline{1}$11	5.87	6	5.87	5.87	9
201	4.25	48	4.25	4.23	63
111	3.94	8	3.95	3.94	11
200 $\overline{2}$00	3.88	3	3.89	—	—
130 $\overline{1}$30	3.782	80	3.782	3.784	82
$\overline{1}$31	3.620	8	3.619	3.620	21
$\overline{2}$21	3.552	6	3.554	3.551	18
$\overline{1}$12	3.458	71	3.460	3.461	60
220 $\overline{2}$20	3.339	100	3.339	3.337	100
$\overline{2}$02	3.293	88	3.293	3.289	89
040	3.249	70	3.249	3.250	71
002	3.228	85	3.228	3.228	91
131	2.993	65	2.995	2.995	60
$\overline{2}$22	2.934	7	2.937	—	—
041	2.902	34	2.902	2.904	40
022	2.891	15	2.891	Kalsilite (003)[1]	
$\overline{1}$32	2.765	13	2.764	2.766	25
$\overline{3}$12	2.608	8	2.606	2.606	24
$\overline{2}$41	2.588	32	2.586	2.589	60
112	2.555	10	2.554	—	—
310 $\overline{3}$10	2.548	13	2.545	—	—

[1] Ill-defined peaks by association with kalsilite.

TABLE 2—(*continued*)

hkl	Sanidine synthesized from KAlSi$_3$O$_8$ glass (20 kb, 600° C.) (this paper)			Sanidine formed by breakdown of leucite (L.97, 29 kb, 700° C.) (this paper)	
	Measured		Calculated	d(Å)	I
	d(Å)	I	d(Å)		
150 $\}$ $\bar{1}50$	2.465	4	2.465	Kalsilite (111)[1]	
$\bar{1}51$	2.418	6	2.418	Kalsilite (103, 013)[1]	
$\bar{2}42$	2.316	8	2.314	2.315	12
042	2.290	8	2.290	2.290	10
$\bar{3}32$	2.276	15	2.276	2.275	16
$\bar{2}23$ $\}$ 132	2.231	14	2.231 22.33	2.231	12
151	2.204	4	2.204	2.204	12
060 $\}$ $\bar{3}13$	2.167	27	2.166 2.165	Kalsilite (201)[1]	
311	2.083	6	2.082	2.079	10
$\bar{1}33$	2.071	6	2.070	2.069	6
222	1.975	25	1.978	1.976	20
$\bar{2}61$ $\}$ $\bar{3}51$	1.930	13	1.931	1.929	13
$\bar{2}43$	1.917	6	1.917	1.918	14
113	1.855	6	1.855	1.853	10
350	1.835	6	1.836	—	—
043	1.794	24	1.794	1.796	30
$\bar{1}53$	1.747	6	1.746	1.746	7
$\bar{3}34$	1.632	11	1.631	—	—

$a = 8.66$ (2) Å	$a = 8.66$ (2) Å
$b = 13.00$ (0) Å	$b = 13.01$ (0) Å
$c = 7.18$ (3) Å	$c = 7.18$ (3) Å
$= 116°$	$= 116°$
$C\ 2/m$	$C\ 2/m$
unit-cell	unit-cell
volume $= 181.7$ Å³	volume $= 181.8$ Å³

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TABLE 3. X-ray Powder Data, Unit-Cell Dimensions and
Unit-Cell Volume, of Hexagonal Kalsilites

hkl	Hexagonal kalsilite formed by breakdown of leucite (29 kb, 700°) (this paper)			Synthesized kalsilite Ne:Ks=0:100 (Smith & Tuttle, 1957)	
	Measured		Calculated	d(Å)	I
	d(Å)	I	d(Å)		
002	4.34	25	4.34	4.3512	12
011 101	3.97	48	3.97	3.9733	45
102 012	3.117	100	3.116	3.1184	100
003	2.899	18	2.899	—	—
110	2.579	38	2.580	2.5789	50
111	2.473	21	2.473	2.4724	15
103 013	2.431	15	2.432	2.4319	10
020 200	2.236	12	2.236	2.236	3
112	2.218	10	2.218	2.2183	10
004	2.176	21	2.176	2.1753	17
201 021	2.166	20	2.166	2.164	5
202 022	2.988	11	1.989	1.9868	5
104 014	1.955	12	1.956	1.9546	3
113	1.929	8	1.929	1.9270	4
023 203	1.772	9	1.771	1.7703	3
114	1.662	5	1.662	1.6621	4
211 121	1.659	11	1.659	1.6580	4

TABLE 3—(*continued*)

hkl	Hexagonal kalsilite formed by breakdown of leucite (29 kb, 700°) (this paper)			Synthesized kalsilite Ne:Ks=0:100 (Smith & Tuttle, 1957)	
	Measured		Calculated	d(Å)	I
	d(Å)	I	d(Å)		
015⎫ 105⎭	1.622	6	1.624	1.6220	6
212	1.567	15	1.567	1.5742	9
204	1.558	8	1.558	1.5588	3
300⎫ 030⎭	1.490	17	1.492	1.4895	10

$a = 5.16$ (4) Å
$c = 8.70$ (5) Å

Hexagonal
Unit-cell volume = 100.5 Å³

2θCuKα (102) -2θCuKα (101) = 6.25°

$a = 5.159$ (7) Å⎫ ± 0.002 Å
$c = 8.703$ (2) Å⎭

Hexagonal
Unit-cell volume
= 100.3 Å³

2θCuKα (102) -2θCuKα
(101) = 6.26°

due to unmixing of a "soda-leucite" (Larsen and Buie, 1938; Larsen *et al.*, 1939). However, Bowen and Ellestad (1937) suggested that leucite can contain no more than 1–1.5% by weight Na$_2$O. The average Na:K ratios for leucite from Africa (Sahama, 1952) and for leucite from Vesuvius (Tschirwinsky, 1931) are 8.5:91.5 and 15.4:84.6 respectively. Thus, leucite is believed to be generally very poor in soda.

The pseudoleucite is also believed to be derived from unmixing breakdown of a potash-analcite into potash-feldspar and nepheline (Fudali, 1957, 1963) Larsen and Buie, 1938; Larsen *et al.*, 1939, 1941; Tilley, 1957). Larsen and Buie (1938) have given the chemical analysis of a potash-rich analcite (Na$_2$O = 8.48%, K$_2$O = 4.48% wt) which formed a primary phenocryst in basaltic rock of the Highwood Mountains in Montana. Barrer (1950) concluded from his experimental work that synthesized analcite may readily exchange Na for K in KCl-bearing aqueous solution. However, potash-rich analcite is very rare, and most natural analcites are very rich in soda and have only minor amounts of K$_2$O.

The hypothesis of chemical reaction between leucite crystals and magmatic or hydrothermal residual solutions which was originally presented

by Bowen and Ellestad (1937) has recently been supported with excellent evidence by Zies and Chayes (1960). Pseudoleucite chiefly composed of potash-feldspar and nepheline with minor kalsilite in solid solution may be fully explained by the reaction theory.

The results of our experimental work as described above, on the other

TABLE 4. X-RAY POWDER PATTERN, UNIT-CELL DIMENSIONS, UNIT-CELL VOLUME, DENSITY AND REFRACTIVE INDEX OF $KAlSi_3O_8 \cdot H_2O$ FORMED BY THE BREAKDOWN OF LEUCITE

hkl	d(Å)	I	hkl	d(Å)	I
001	7.67	4	004	Kalsilite (113)[1]	
010⎫ 100⎭	4.61	28	113	1.847	10
101⎫ 011⎬ 002⎭	Kalsilite (011)[1]		120⎫ 210⎭	1.746	2
102⎫ 012⎭	2.956	100	023⎫ 203⎭	1.714	4
110	2.665	74	211⎫ 121⎭	1.700	4
111	2.514	7	122⎫ 212⎭	1.591	15
200⎫ 020⎭	2.307	15	114	Kalsilite (212)[1]	
103	Kalsilite (020)[1]		005⎫ 300⎬ 030⎭	1.540	12
201⎫ 021⎭	Kalsilite (112)[1]				

Hexagonal
  $a = 5.33$ Å
  $c = 7.70$ Å
  unit-cell volume $= 189$ Å³
  calculated density $= 2.60$
  $n = 1.54$
[1] Ill-defined peaks by association with kalsilite.

hand, are believed to show that the unmixing of leucite during the cooling stage of a magma may be also a very important process in the formation of pseudoleucite. From our data it is clear that pure leucite must be broken down into a mixture of sanidine and hexagonal kalsilite with increasing pressure and/or decreasing temperature by the following chemical reaction:

	2 leucite	$\longrightarrow$	sanidine	$+$	hexagonal kalsilite
	$2KAlSi_2O_6$		$KAlSi_3O_8$		$KAlSiO_4$
molecular volume	146.7 Å³		181.8 Å³		100.5 Å³
total volume	293.4 Å³			282.3 Å³	

From the solid volume relations shown above (see also Table 5) increasing pressure will strongly favor the breakdown of leucite.

Figure 1 also shows that this chemical reaction will be favored by falling temperatures.

Yagi (1954) stressed the fact that pseudoleucite of the Tzu Chin Shan district of China is low in soda and high in potash and is chiefly composed of sanidine and kalsilite (potash-rich nepheline). He presents such excel-

TABLE 5. PHYSICAL PROPERTIES OF LEUCITE, SANIDINE,
HEXAGONAL KALSILITE AND $KAlSi_3O_8 \cdot H_2O$

	Leucite	Sanidine	Hexagonal Kalsilite	$KAlSi_3O_8 \cdot H_2O$
Density	2.47	2.57	2.59	2.60
Refractive index	5.508–1.509	1.520–1.53	1.537–1.542	1.54
Hardness	5.5–6	6	6	n.d.
Packing index	4.7	4.9—5.0	4.93	5.24

lent evidence that it is preferable to infer that the pseudoleucite was formed by unmixing of leucite rather than by reaction of the original leucite with a sodic residual magma.

It is certainly true that most pseudoleucite-bearing rocks have been hydrothermally altered. For example olivine crystals in pseudoleucite-bearing lavas were almost completely altered to chlorite or other ferromagnesian hydrous minerals (Balsillie, 1936; Prider and Cole, 1942; Wade and Prider, 1940). On the other hand, when the leucite-bearing rocks are fresh, no transformation of leucite into potash-feldspar-bearing mineral assemblages can be found (Davis and Fisset, 1947; Holmes, 1945; Sahama, 1952).

We have noted in our work that leucite never alters into another phase or phases under dry conditions even in the long, high temperature and high pressure experimental runs. Barrer and Hinds (1950) and Barrer, Hinds and White (1953) reported that synthesized pure leucite in the aqueous solution of K_2CO_3 and Na_2CO_3 was broken into a mixture containing orthoclase after 6 hours at 195–200° C. in an autoclave. They also succeeded in converting pure leucite into kalsilite by keeping the leucite

saturated with 3N-KOH solution at 250–350° C. for 2 or 3 days in the autoclave.

Thus, it is quite possible that the presence of water, aqueous solution or vapor is necessary for the formation of pseudoleucite.

Throughout this paper we have treated our experimental results as though total pressure and partial pressure of water were the same. This assumption is of no importance in locating the boundary between the leucite—sanidine plus kalsilite field, but it is of importance in locating the boundary between the fields of sanidine and $KalSi_3O_8 \cdot H_2O$. Here the partial pressure of water is certainly not the total pressure on the system as undoubtedly large and nonstoichiometric amounts of potash, alumina and silica are dissolved in the water, thus changing the chemical potential and partial pressure of the water. We show the boundary, thus, for our experimental conditions with relatively "small" amounts of fluid at high pressures.

ACKNOWLEDGEMENTS

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