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Phase Transition in Synthetic and Natural Leucite¹⁾

By *George T. Faust* (Washington)

With 3 figures in the text and 9 tables

Abstract

The polymorphic inversion of leucite was investigated by differential thermal analysis. The inversion is reversible but is not isothermal. It consists of two processes. The first is rapid and yields a sharp minimum; the second yields a broad and rounded minimum. These data suggest the existence of either a transient polymorphic phase or an Al-Si order-disorder relationship. This matter is under investigation. X-ray powder diffraction data are given for synthetic leucite, six naturally occurring leucites, and a synthetic iron-leucite. These data were refined by a recently developed least squares method for the refinement of unit cell parameters using X-ray powder data from which calculations the corresponding hkl and unit cell dimensions were obtained.

The significance of the polymorphic inversion in petrogenetic theory is discussed. Leucite crystallizes in magmas at temperatures probably in excess of 700°C. The size of the crystals and their skeletal development suggest that leucite grows rather quickly and in a fluid melt.

Zusammenfassung

Die polymorphe Umwandlung des Leucits wurde unter Anwendung der Differentialthermo-Analyse untersucht. Die Umwandlung ist umkehrbar; aber sie ist nicht isothermal. Sie besteht aus zwei Vorgängen. Der erste ist schnell und ergibt ein scharfes Minimum; der zweite Vorgang ergibt ein breites und abgerundetes Minimum. Diese Daten deuten auf das Vorhandensein einer vorübergehenden polymorphen Phase oder auf eine Al-Si-Ordnung-Unordnung-Beziehung. Die Röntgendiffertenzdaten von Pulverdiagrammen für einen synthetischen Leucit, sechs in der Natur vorkommende Leucite und einen synthetischen Eisen-Leucit werden angegeben. Diese Daten wurden mittels einer kürzlich entwickelten „kleinsten

¹⁾ Contribution from the Mineralogical Laboratory of the University of Michigan and the U.S. Geological Survey. Publication authorized by the Director, U.S. Geological Survey.

Quadrat"-Methode verfeinert; eine Methode zur Verfeinerung von Gitterkonstanten von Elementarzellen, welche Pulverdaten verwendet. Auf Grund dieser Berechnungen wurden die entsprechenden hkl und Zelldimensionen erhalten.

Die Bedeutung der polymorphen Umwandlung in der Theorie der Petrogenesis wird erörtert. Leucit kristallisiert im Magma bei einer Temperatur von wahrscheinlich über 700°C . Die Grösse der Kristalle und die Entwicklung ihrer Skelette weisen darauf hin, dass Leucite ziemlich schnell wachsen, und das in einer flüssigen Schmelze.

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Statement of Progress

While in the Mineralogical Laboratory of the University of Michigan (1930—1935) I studied the system $\text{K}_2\text{O}-\text{Fe}_2\text{O}_3-\text{SiO}_2$. Quite unexpectedly, I found that the liquidus surfaces, for compositions on the join $\text{K}_2\text{O}\cdot 6\text{SiO}_2-\text{Fe}_2\text{O}_3$ containing approximately 10 to 18 per cent ferric oxide, were at unusually low temperatures. These temperatures range from about 900°C to as low as 740°C . Similar relationships were observed along other joins and tie-lines in the system. Those compositions contained within the low temperature portions of the liquidus surfaces when prepared as homogenous glasses exhibited a very great viscosity. As a result of these conditions some of the liquids failed to develop crystals when held at appropriate temperatures for periods of time as long as two years. In marked contrast to this behavior a melt of the composition of iron-leucite ($\text{K}_2\text{O}\cdot\text{Fe}_2\text{O}_3\cdot 4\text{SiO}_2$) could be completely

crystallized. Melts of the composition of iron-orthoclase ($K_2O \cdot Fe_2O_3 \cdot 6SiO_2$) when crystallized yielded mixtures of crystals of iron-orthoclase and glass and the iron-orthoclase melted incongruently to yield iron-leucite and a liquid.

The results of some of this experimental work have been published. FAUST (1936) described the fusion relations of iron-orthoclase and presented the phase relationships along the join $K_2O \cdot 6SiO_2$ (glass)- Fe_2O_3 . FAUST and PECK (1938) described the chemical and optical properties of the glasses in the sub-system $K_2O \cdot 4SiO_2 - SiO_2 - Fe_2O_3$. Additional studies were planned but not completed at that time. These were the complete description of the phase relations in the system $K_2O \cdot 4SiO_2 - SiO_2 - Fe_2O_3$ and the descriptions of the physical properties of the phases iron-leucite and iron-orthoclase. This system is still under investigation and the results of the study of the phase, iron-leucite in comparison with leucite, are presented here.

Previous Studies of the Inversion Phenomena in Leucite and Iron-Leucite

The discovery of the inversion of low leucite to high leucite was made many years ago as a result of the studies of the optical properties of anomalously birefringent crystals. CARL KLEIN (1884a, 1884b, 1897) summarized the early work and notably extended the thermo-optical studies. KLEIN concluded from his work that the natural mineral leucite crystallized in magmas as cubic crystals and subsequently acquired its supposed orthorhombic symmetry on cooling. The basis for the symmetry determination of the high leucite as cubic was the isotropic character of the high temperature phase and the crystal morphology (tetragonal trisoctahedra). Low leucite was judged to be orthorhombic on the basis of VOM RATH's (1872) study of the twinning striae and morphology and BAUMHAUER's (1877) examination of the etching phenomena. The early thermo-optical studies on the inversion of leucite placed the value for the inversion point at temperatures within the range of 433 to 560°. This variation was due to the poor quality of the thermometric measurements. Later studies by RINNE and KOLB (1910) reported that the birefringence of a transparent crystal, from the tuff at Campagna near Rome, Italy, decreased rapidly at 684° C and disappeared at 714° C.

GROSSMANN (1917) using a thermo-goniometric method measured the inversion temperature of leucite from Vesuvius, Italy at 620° C. BOWEN and SCHAIER (1929) using the method of cooling and heating curves

placed the inversion of pure synthetic leucite at 603° C but indicated that they planned to investigate the inversion relationships in more detail. They observed that this inversion was instantaneously reversible. WYART (1938) using the DTA method observed what he termed a sinuosity in the curve between 575° and 625° and this he observed was repeated on cooling confirming the earlier studies on the reversible character of the inversion. BERKELHAMER (1944) in a search for possible interferences in the region of the quartz inversion made a DTA study of leucite crystals from Alban Hills, Rome, Italy and found the inversion to be a double break and to take place over an interval. He made no comments on the results of this study. KIRSCH (1956) made heating and cooling curve studies on leucite using a DTA apparatus. He noted the reversible nature of the transformation and observed only one break in the curve. He also studied a leucite basanite bomb, from Vesuvius, with similar but subdued effects. McLAUGHLIN (1957) studied leucite from Vesuvius by DTA and ascribed the double peak "to the presence of particles of different particle sizes".

It is noteworthy that the determination of the inversion temperature by the diverse technique failed to yield an unequivocal inversion point but indicated that the inversion took place rather slowly and was not abrupt in character like the low-high quartz inversion. Thus RINNE and KOLB (1910) point out that the birefringence began to diminish at 684° before disappearing at 714° when the specimen becomes isotropic. They noted that the inversion was not sharp and that it spread out over an interval of 30° C. They suggested that the inversion interval might be due to solid solution with the sodium analog of leucite. GROSSMANN's (1917) study of the face angles of leucite as a function of temperature shows some rounding of the curve at 600°. CHEVENARD and PORTVEIN (1943) studied this inversion with a dilatometer and observed that the transformation of 625° C is "almost perfectly reversible but not exactly isothermal". BERKELHAMER (1944) did not discuss the appearance of two troughs on the curve for leucite. His DTA curve shows two rounded troughs with minima at 650° C and about 675° C. The results of the previous measurements, interpreted in part by FAUST, are given in Table 1.

The type of inversion phenomena considered here is associated with those crystals characterized as having a mimetic crystal habit. Leucite and boracite ($Mg_7B_{16}O_{30}Cl_2$) are good examples. The inversion in a cooling process gives rise to the complicated twinning phenomena in leucite consisting of intersecting systems of twin lamellae sometimes

Table 1. *Inversion temperature for leucite as determined by others*

| Year | Method | Low→ high °C | High→ low °C | Inversion interval, °C | Investigator |
|---------------|--|--------------------------------------|-----------------|-----------------------------|---------------------------|
| Up to 1897 | Optical | 433 to 560 | — | — | Various |
| 1910 | Optical Interval | 684—714 | — | 30 | RINNE and KOLB |
| 1917 | Thermo- gonio- metric | 620 | — | 20 (Estimated G.T.F.) | GROSSMAN |
| 1929 | Thermal analysis by cooling curve | 603 | — | — | BOWEN and SCHAIRER |
| 1938 | DTA | 575—625 | Noted | 50 | WYART |
| 1943 | Dilatometer | 625 | Noted | Noted | CHEVENARD and PORTVEIN |
| 1944 | DTA | 650 and 675 (Estimated G.T.F.) | — | 25 | BERKELHAMER |
| 1956 | DTA | 620 | Noted | — | KIRSCH |
| 1957 | DTA | 611 and 633 | — | 22 | McLAUGHLIN |

described as organ-pipe structure (WEATHERHEAD, A. V., 1945, Photomicrographs of rock-forming minerals: Geologist's Assoc., Pr. v. 56, pt. 1, p. 17).

The occurrence of the inversion in leucite and the presence of the twinning phenomena in both leucite and iron-leucite suggested that a parallel thermal relationship should exist in iron-leucite. The present study was undertaken to verify this suggestion and to re-examine leucite.

Descriptions and Chemical Analyses of the Specimens Studied

The synthetic leucites used in this study were prepared from purified reagents by BOWEN and SCHAIRER (1929), in their investigations of the binary system leucite-diopside. Dr. J. F. SCHAIRER of the Geophysical Laboratory very kindly made available the samples listed below:

Leucite No. 1. Synthetic preparation, crystallized at 1200° C for 4 days. $\text{Na}_2\text{O} = 0.10$, $\text{CaO} = 0.04$ by flame photometer. J. Dinnin, Analyst.

Leucite No. 2. Synthetic preparation, no. 400 1/2, leucite 95%, diopside 5%. Crystallized at 1200° C.

Leucite No. 3. Synthetic preparation, no. 402, leucite, 80%, diopside 20%. Crystallized at 1200° C for 25 hours.

Leucite No. 4. Synthetic preparation, no. 405, leucite 50%. diopside 50%. Crystallized at 1200° for 18 hours.

The natural leucites studied are chiefly from Italian rocks. The localities given below have been described by HENRY S. WASHINGTON (1906, 1920, 1927) who collected some of the specimens.

Leucite No. 5. Locality: Villa Senni near Grotto Ferrata, on the north slope of the Alban Hills, Italy. Collected by H. S. Washington. Frosted-like tetragonal trisoctahedrons. Crystals about 1 cm — about the girth. U.S.N.M. No. 96430. Optics by H. E. MERWIN in WASHINGTON (1927, p. 175).

Leucite No. 6. Locality: Monte Somma, Vesuvius, Italy. Frosted-like tetragonal trisoctahedral crystal of about 1.7 cm about the girth. From a leucite basanite U.S.B.M. No. 36738.

Leucite No. 7. Locality: Villa Senni near Grotto Ferrata, Alban Hills, Italy. Collected by H. S. Washington. Frosted-like tetragonal trisoctahedrons crystals about 0.8 mm about the girth. U.S.N.M. No. 95709.

Leucite No. 8. Locality: Vesuvius, Italy. Tetroganal trisoctahedron. U.S.N.M. No. R 2946.

Leucite No. 9. Locality: Garofali, Roccamontfina, Italy. Fragment of a large tetragonal trisoctahedron having a girth of about 3 cm. U.S.N.M. No. 102805.

Leucite No. 10. Locality: From the wall of a building in Pompeii, Italy. Collected in 1912 by Dr. Waldemar T. Schaller.

Leucite T.G.S. 1. Locality: Nyiragongo, NE of Lake Kivu, Republic of the Congo. Specimen collected by Professor T. G. Sahama and numbered by him leucite FEAE-88. The analysis given in Table 2 is reproduced by the generosity of Professor Sahama.

Leucite T.G.S. 2. Locality: Flow from Mt. Mikeno, 1 mile south of Rulenga Mission, Lake Kivu, National Park Area, Republic of the Congo. From a specimen of mikenite (a leucite of Mikeno type). The analysis in Table 2 is taken from SAHAMA (1952).

Leucite T.G.S. 3. Locality: Villa Senni, Grotto Ferrata, Alban Hills, Italy. Portion of an analyzed sample (see Table 2) kindly sent to the author by Professor Sahama.

Iron-leucite. Synthetic preparation, prepared from purified reagents, synthesized by FAUST (1936).

Table 2. Chemical analyses of the leucites

| Number | 5 | 6 | 7 | 8 | 9 | 10 | T.G.S.-1 | T.G.S.-2 | T.G.S.-3 |
|--|---|------------------------------|---|-----------------|-------------------------------|----------------|---------------------------------|---|--|
| Mineral | Leucite | Leucite | Leucite | Leucite | Leucite | Leucite | Leucite | Leucite | Leucite |
| Locality | Villa Senni, Grotto Ferrata, Alban Hills, Italy | Monte Somma, Vesuvius, Italy | Villa Senni, Grotto Ferrata, Alban Hills, Italy | Vesuvius, Italy | Garofali, Roccamonfina, Italy | Pompeii, Italy | Nyiragongo, The Congo (FEAE-85) | From Mt. Miken, Rulenga Mission, The Congo (C-2912) | Villa Senni, Grotto, Ferrata, Alban Hills, Italy |
| SiO ₂ | | | | | | | 54.80 | 54.62 | 54.60 |
| Al ₂ O ₃ | 21.1 | 20.0 | 21.1 | 19.4 | 19.8 | 21.0 | 23.34 | 22.93 | 21.97 |
| K ₂ O ¹⁾ | 0.28 | 1.04 | 0.27 | 1.24 | 0.95 | 0.26 | 0.95 | 21.02 | 21.45 |
| Na ₂ O ¹⁾ | 0.12 | 0.14 | 0.12 | 0.14 | 0.14 | 0.16 | 0.09 | 0.08 | 0.23 |
| CaO ¹⁾ | | | | | | | 0.04 | 0.00 | 0.10 |
| MgO | | | | | | | 0.19 | 0.00 | 0.00 |
| TiO ₂ | | | | | | | 0.53 | 0.26 | 0.89 |
| Fe ₂ O ₃ ²⁾ | 0.43 | 0.29 | 0.43 | 0.29 | 0.71 | 0.14 | 0.00 | 0.26 | 0.14 |
| FeO | | | | | | | | 0.00 | 0.00 |
| MnO | | | | | | | | | 0.00 |
| | | | | | | | 100.29 ³⁾ | 99.95 ⁴⁾ | 99.54 ⁵⁾ |

¹⁾ Determined by the flame spectrometer. Analysts: PAUL SCOTT, PAUL L. D. ELMORE, and JOSEPH I. DINNIN.²⁾ Determined spectroscopically as Fe, calculated as Fe₂O₃. Analytical error \pm 0.07% Fe₂O₃. Analyst: HARRY BASTRON.³⁾ Others: -CO₂ = 0.00; H₂O(+) = 0.34; H₂O(-) = 0.00. Analyst: H. B. WINK.⁴⁾ Others: -H₂O(+) = 0.12; H₂O(-) = 0.00. Analyst: H. B. WINK.⁵⁾ Others: -H₂O(+) = 0.13; H₂O(-) = 0.00; CO₂ = n. d. Analyst: T. G. SAHAMA.

Table 2 contains the chemical analysis of the natural leucites. The partial analyses were made to determine the extent of solid solution relationships. The partial analyses were made on the material removed from the nickel block of the DTA apparatus after the completion of the thermal studies.

X-ray Studies on Leucite and Iron-Leucite X-ray Powder Diffraction Data

X-ray powder patterns of synthetic iron-leucite and natural leucite from Villa Senni, Grotto Ferrata, Alban Hills, Latian district, Italy, were made at the Mineralogical Laboratory of the University of Michigan in 1934. The patterns were prepared using molybdenum radiation, filtered by zirconium, in cassettes mounted on a General Company apparatus of the type described by DAVEY (1922). The patterns obtained although entirely satisfactory for proving the identity of the synthetic iron-leucite with natural leucite were made up of lines too broad for the measurement of reliable interplanar spacings. Good patterns of these leucites and other leucites were prepared at the U. S. Geological Survey using copper radiation with a nickel filter and chromium radiation with a vanadium filter. The chromium radiation provided patterns of unusual clarity and were used for measurement of the interplanar spacings. The powder patterns were taken with a Debye-Scherrer camera (114.59 mm diameter) using the Straumanis technique and vanadium-filtered $\text{CrK}\alpha$ radiation ($\lambda = 2.2909 \text{ \AA}$). The films were corrected for shrinkage.

The results of the measurements on synthetic iron-leucite, leucite and six natural leucites are given in Table 3 as d (Obs.). Originally, the x-ray powder diffraction data for the natural leucite from Pompeii, Italy (Specimen No. 10) was indexed by conventional methods. The development of the program for the refinement of unit cell parameters from x-ray powder data, using the method of least squares, by Howard T. Evans, Daniel Appleman and David Handwerker of the U. S. Geological Survey, has enabled the dimensions of the unit cells to be determined quite accurately. These data, very kindly computed by my colleague Howard T. Evans, on a Burroughs B220 digital computer, are given in Table 3. These computations were based on the space group for leucite deduced by WYART (1937, 1940) and confirmed by VON NÁRAY-SZABÓ (1942), namely, $\text{C}_{4h}^6 - \text{I}4_{1/a}$. The intensity relations of the lines in the powder patterns of the natural leucites are

Table 3. X-ray powder data for natural leucite, synthetic leucite, and iron-leucite

| Garofali, Rocca-montina, Italy | Nyiragongo, The Congo | Villa Senni, Alban Hills, Italy | Vesuvius Italy | Pompeii Italy | Monte Somma Italy | I $\sqrt{2}$ scale | Synthetic leucite I $\sqrt{2}$ scale | Synthetic iron-leucite I $\sqrt{2}$ scale |
|--------------------------------|-----------------------|---------------------------------|--------------------|--------------------|--------------------|--------------------|--------------------------------------|---|
| hkl | d (Calc.) d (Obs.) | d (Calc.) d (Obs.) | d (Calc.) d (Obs.) | d (Calc.) d (Obs.) | d (Calc.) d (Obs.) | d (Calc.) d (Obs.) | d (Calc.) d (Obs.) | d (Calc.) d (Obs.) |
| 101 | 9.478 9.493 | 9.471 9.493 | 9.470 9.480 | 9.460 9.493 | 9.458 9.439 | 9.452 9.412 | 6 9.472 9.535 | 6 9.596 9.623 |
| 200 | 6.540 6.551 | 6.530 6.596 R | 6.525 6.564 R | 6.523 6.487 | 6.507 5.496 | 5.502 5.514 | 2 5.514 5.509 | 7 5.593 5.625 |
| 112 | 5.518 5.527 | 5.515 5.541 R | 5.515 5.541 | 5.376 5.394 | 5.371 5.364 | 5.371 5.377 | 7 5.370 5.360 | 11 5.380 5.386 |
| 211 | 5.383 5.394 | 5.377 5.364 | 5.376 5.394 | 4.735 4.748 | 4.730 4.738 | 4.729 4.731 | 6 4.726 4.725 | 6 4.736 4.755 |
| 202 | 4.739 4.745 | 4.735 4.738 | 4.735 4.748 | 4.730 4.738 | 4.729 4.731 | 4.726 4.725 | 6 4.736 4.755 | 6 4.798 4.788 |
| 103 | | | 4.325 4.335 | | 4.318 4.292 R | 4.313 4.330 | 3 4.322 4.349 R | 3 4.381 4.385 |
| 222 | 3.837 3.776 R | 3.833 3.784 R | 3.830 3.776 R | 3.829 3.776 R | 3.827 3.774 R | 4 3.835 3.778 R | 4 3.881 3.835 | 4 3.881 3.835 |
| 213 | 3.608 3.602 | 3.606 3.608 | 3.606 3.604 | 3.602 3.599 | 3.601 3.602 | 3.598 3.595 | 6 3.605 3.604 | 6 3.657 3.646 |
| 312 | | | | | | | 3.543 3.525 R | 5 |
| 321 | 3.508 3.511 | 3.503 3.508 | 3.503 3.518 R | 3.500 3.506 | 3.500 3.502 | 3.499 3.504 | 5 | |
| 004 | 3.438 3.442 | 3.438 3.438 | 3.438 3.438 | 3.434 3.433 | 3.432 3.433 | 3.427 3.426 | 12 3.435 3.442 | 12 3.492 3.487 |
| 400 | 3.270 3.275 | 3.266 3.268 | 3.265 3.266 | 3.262 3.268 | 3.262 3.256 | 3.262 3.262 | 14 3.269 3.272 | 13 3.301 3.303 |
| 114 | 3.223 3.218 | | 3.222 3.217 | 3.218 3.218 | | 3.213 3.205 | 1 | |
| 303 | 3.159 3.166 | | 3.157 3.159 | 3.153 3.170 R | | 3.151 3.143 | 1 | |
| 411 | 3.091 3.091 | 3.087 3.091 | 3.087 3.090 | 3.084 3.087 | | 3.090 3.095 | 5 | 3.122 3.118 |
| 204 | 3.043 3.041 | 3.042 3.049 R | 3.042 3.043 | 3.039 3.041 | | 3.034 3.038 | 4 | 3.041 3.049 |
| 420 | 2.925 2.922 | 2.921 2.920 | 2.920 2.918 | 2.918 2.918 | 2.918 2.922 | 2.918 2.916 | 10 2.924 2.922 | 10 2.953 2.950 |
| 323 | 2.845 2.845 | 2.842 2.843 | 2.842 2.842 | 2.839 2.843 | 2.839 2.838 | 2.837 2.827 | 10 2.843 2.843 | 10 2.879 2.878 |
| 332 | 2.813 2.812 | 2.810 2.810 | 2.810 2.808 | 2.807 2.810 | 2.807 2.807 | 2.806 2.804 | 8 2.812 2.810 | 8 2.843 2.837 |
| 422 | | | 2.688 2.685 R | | | 1 | | |
| 105 | 2.691 2.693 R | | | | 2.683 2.683 R | 2.680 2.689 R | 7 | |
| 314 | 2.644 2.644 | 2.642 2.640 | 2.642 2.641 | 2.639 2.639 | 2.639 2.640 | 2.637 2.639 | 6 2.642 2.645 | 6 2.679 2.679 |
| 413 | 2.609 2.604 R | 2.606 2.602 R | 2.606 2.602 | 2.603 2.604 | 2.603 2.596 | 2.602 2.599 | 3 2.607 2.606 | 3 2.639 2.638 |
| 215 | 2.489 2.489 | 2.488 2.487 | 2.488 2.487 | 2.485 2.487 | 2.484 2.490 | 2.482 2.482 | 4 2.487 2.483 | 4 2.526 2.521 |
| 404 | 2.369 2.369 | 2.368 2.368 | 2.367 2.366 | 2.365 2.367 | 2.365 2.369 | 2.363 2.363 | 9 2.368 2.368 | 8 2.399 2.398 |
| 305 | | | 2.325 2.329 | | | 4 | | |
| 440 | 2.312 2.316 R | 2.309 2.308 | 2.309 2.308 R | 2.307 2.310 | 2.307 2.310 | 2.306 2.313 | 4 2.311 2.313 | 4 2.334 2.335 |
| 503 | 2.272 2.271 | | 2.269 2.270 R | 2.267 2.269 | 2.267 2.268 R | 2.266 2.268 | 1 2.268 2.187 R | 1 |
| 442 | 2.192 2.189 R | 2.189 2.188 | 2.190 2.192 R | 2.188 2.189 R | | 1 | | |
| 325 | | | | | | | | |
| 600 | | | | | | | 2.201 2.199 R | 6 |
| 206 | 2.163 2.163 | 2.163 2.162 | 2.163 2.162 | 2.160 2.160 | 2.159 2.162 | 2.156 2.160 | 4 2.161 2.159 | 4 2.154 2.156 |
| 532 | 2.133 2.130 R | 2.130 2.129 | 2.130 2.127 | 2.128 2.129 | 2.128 2.128 | 2.127 2.127 | 7 2.132 2.132 | 6 2.134 2.136 |
| 620 | 2.068 2.066 R | 2.065 2.064 | 2.065 2.065 | 2.063 2.064 | 2.063 2.066 | 2.063 2.064 | 4 2.067 2.064 | 4 2.084 2.084 |
| 226 | | | | | | | | |
| 541 | 2.021 2.020 | | 2.018 2.012 R | 2.016 2.014 | 2.016 2.016 | 2.016 2.015 | 3 2.020 2.020 | 3 2.040 2.039 |
| 622 | | 1.978 1.980 | | | | 1 | | |
| 613 | | 1.943 1.942 | 1.943 1.943 R | 1.940 1.941 | 1.942 1.941 R | 1.942 1.939 R | 3 1.941 1.942 | 3 1.949 1.945 R |
| 107 | 1.943 1.942 | | | | | | | |
| 631 | | | | | | | | |
| 444 | 1.919 1.920 | | 1.917 1.919 | 1.915 1.918 R | 1.915 1.918 | 1.914 1.914 | 3 1.918 1.918 | 3 1.968 1.969 |
| 534 | | 1.877 1.877 R | 1.876 1.874 | 1.877 1.877 R | 1.875 1.875 R | 1.872 1.873 R | 3 1.875 1.874 | 3 1.903 1.904 |
| 406 | 1.877 1.873 R | 1.876 1.874 | | 1.862 1.862 | 1.862 1.861 | 1.861 1.861 | 3 1.865 1.865 | 3 1.891 1.889 |
| 543 | 1.866 1.866 | 1.864 1.865 | | | | | | |
| 217 | | | | | | | | |
| 604 | | | | | 1.836 1.836 | 1.840 1.840 R | 3 | 1.864 1.866 |
| 336 | 1.839 1.837 R | 1.838 1.838 R | 1.838 1.837 R | 1.836 1.837 R | 1.836 1.834 R | 1.810 1.807 | 2 1.813 1.814 | 2 1.831 1.833 |
| 640 | 1.814 1.815 | | | | | | 3 1.790 1.790 | 3 1.813 1.814 |
| 633 | | | | | | | | |
| 307 | | 1.791 1.789 | | 1.789 1.786 R | 1.788 1.788 R | | | |
| 712 | 1.786 1.787 | | 1.784 1.785 | | | 3 | | 1.804 1.804 |
| 721 | | | 1.777 1.780 R | | | | 1.781 1.783 | 3 |
| 642 | | | 1.750 1.743 R | | | | | 1.748 1.750 R |
| 703 | | 1.728 1.728 R | 1.727 1.726 R | | 1.726 1.727 R | 1.726 1.725 | 6 1.726 1.728 R | 6 1.748 1.750 R |
| 327 | 1.727 1.728 | | | | | | | |
| 723 | | | | | | | | |
| 417 | 1.670 1.667 R | 1.664 1.663 R | 1.668 1.667 | 1.667 1.666 | 1.669 1.668 | 3 1.669 1.667 R | 8 1.690 1.691 | 5 |
| 732 | 1.666 1.668 R | 1.662 1.663 | 1.660 1.665 R | 1.631 1.628 | | | | 1.683 1.683 |
| 208 | | | | | | | | |
| 800 | | | | | | | | |
| 714 | 1.627 1.627 R | 1.627 1.629 R | 1.626 1.625 R | | 1.625 1.626 | 6 1.627 1.627 R | 5 1.648 1.649 R | 5 |
| 446 | | | | | | | | |
| 644 | | | | | | | | |
| 802 | | | | | | | | |
| 318 | | | | | | | | |
| 820 | 1.586 1.586 | 1.584 1.584 | 1.584 1.585 | | | 6 1.586 1.584 | 6 1.601 1.600 R | 3 |
| 653 | 1.571 1.570 | 1.571 1.571 R | 1.571 1.571 R | 1.568 1.568 | 1.569 1.569 R | 1.566 1.568 R | 4 1.570 1.570 R | 4 1.592 1.592 R |
| 507 | | | | | | | 2 1.545 1.555 R | 2 |
| 437 | | | | | | | | |
| 822 | | | | | | | | |
| 734 | | | | | 1.533 1.533 R | 1.533 1.533 | 5 1.534 1.534 | 5 1.554 1.555 R |
| 626 | | | | | 1.526 1.526 R | 1.533 1.532 | | |
| 527 | | | | | | | | |
| 662 | | | | | | | | |
| 338 | | | | | | | | |
| 752 | 1.485 1.484 | | 1.481 1.481 R | 1.480 1.479 | 1.481 1.482 | 1.481 1.480 | 6 1.481 1.481 | 6 1.501 1.500 R |
| 428 | 1.478 1.479 | 1.478 1.480 R | | | | | | |
| 840 | | | | | | | | |
| 309 | | | | 1.440 1.440 | 1.439 1.439 R | | | |
| 824 | | | | | | 4 | | |
| 716 | | | | | | | 1.439 1.438 R | 4 1.457 1.457 R |
| 547 | | | | | 1.415 1.414 | 1.413 1.415 R | 2 1.408 1.408 R | 3 1.434 1.435 |
| 912 | 1.414 1.414 | | | | 1.408 1.408 | | 1.408 1.408 | |
| 921 | | | | | | | | |
| 761 | | | | | | | | |
| 329 | 1.408 1.409 R | | 1.409 1.409 R | | 1.408 1.408 R | | 5 1.407 1.408 | 5 1.421 1.419 R |
| 664 | | | | | | | | |
| 903 | | | | | | | | |
| 637 | | | | | | | | |
| 448 | | | | | | | | |
| 419 | | | | | | | | |
| 736 | 1.374 1.373 R | 1.373 1.374 R | 1.373 1.373 | 1.375 1.373 R | 1.374 1.374 | 1.362 1.361 | 6 1.374 1.373 | 6 1.391 1.391 |
| 538 | | | | | | | | |
| 11.10 | 1.360 1.360 | 1.360 1.361 R | 1.360 1.360 | 1.358 1.359 R | 1.357 1.353 R | 1.361 1.360 | 4 1.359 1.359 | 4 1.382 1.382 |
| 763 | | | | | | | | |
| 707 | | | | | | | | |
| 844 | | | | | | | | |
| 20.10 | | | | | | | | |
| 853 | | | | | | | | |
| 727 | | | | | | | | |
| 941 | | | | | | | | |
| 628 | | | | | | | | |
| 10.00 | | | | | | | | |
| 31.10 | | | | | | | | |
| 826 | 1.304 1.304 | | 1.305 1.305 | 1.305 1.305 | 1.305 1.304 R | 1.305 1.305 | 7 1.304 1.304 R | 7 1.325 1.324 R |
| 772 | | | | | | | | |
| 10.11 | 1.296 1.296 | | 1.294 1.294 | 1.293 1.293 | 1.294 1.294 | | 7 1.295 1.294 R | 7 1.319 1.320 R |
| 943 | | | | | | | | |
| 657 | | | | | | | | |
| 756 | | | | | | | | |
| 10.22 | | | | | | | | |
| 925 | | | | | | | | |
| 718 | 1.259 1.259 | 1.259 1.259 R | | 1.258 1.258 R | | 1.249 1.250 R | | 1.275 1.276 |
| 10.13 | | | | | | | | |
| 848 | | | | | | | | |
| 10.15 | | | | | | | | |
| 828 | | | | | | | | |

¹⁾ Films corrected for shrinkage. Camera diameter 114.59 mm. Chromium radiation filtered by Vanadium ($\lambda = 2.2909$). Lower of 2θ measurable: approximately 4.3° = 15 \AA , B = broad, Temperature 25°C .

The symbol R adjacent to d(obs.) signifies that this particular line was rejected in the least squares refinement but was nevertheless indexed. These are probably ambiguous — should show two or more hkl's.

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nearly identical and accordingly only one set of I values, based on a square root of two scale, are given. This list of intensities was compiled chiefly from the pattern of the leucite from Villa Senni, Alban Hills, Italy but those lacking have been added from the other patterns.

The unit cell parameters a and c and the standard deviations for the natural and synthetic leucites and synthetic iron-leucite, upon which Table 3 is based, are given in Table 4. Similar data for VON NÁRAY-SZABÓ's leucite from Vesuvius is added for comparison.

Table 4. *Unit cell parameters for six natural leucites, a synthetic leucite and iron-leucite measured at 25° C*

| Locality or Origin | a | c |
|--|--------------------|--------------------|
| Garofali, Roccamonfina, Italy | 13.080 \pm 0.001 | 13.752 \pm 0.002 |
| Synthetic leucite | 13.074 \pm 0.003 | 13.738 \pm 0.003 |
| Nyiragongo, The Congo | 13.062 \pm 0.002 | 13.752 \pm 0.002 |
| Villa Senni, Alban Hills, Italy | 13.060 \pm 0.002 | 13.751 \pm 0.003 |
| Vesuvius, Italy | 13.050 \pm 0.001 | 13.734 \pm 0.002 |
| Pompeii, Italy | 13.049 \pm 0.003 | 13.728 \pm 0.006 |
| Monte Somma, Italy | 13.050 \pm 0.002 | 13.709 \pm 0.005 |
| Synthetic iron-leucite | 13.205 \pm 0.002 | 13.970 \pm 0.003 |
| VON NÁRAY SZABÓ's leucite from Vesuvius recalculated in this study | 13.029 \pm 0.002 | 13.847 \pm 0.004 |

VON NÁRAY-SZABÓ (1942) gave full data for his oscillation photographs of a single crystal of leucite from Vesuvius and these were used to derive values of Θ from which the interplanar spacings were calculated and are given as d (Obs.) in Table 5. Using these Θ values in the least squares program for the refinement of unit cell parameters based on x-ray powder data, a second set of interplanar spacings was calculated by Howard T. Evans and is given in Table 5 as d (Calc.). VON NÁRAY-SZABÓ gives as the dimensions of his unit cell the values $a = 13.04 \text{ \AA}$, $c = 13.85 \text{ \AA}$. The dimensions of the unit cell, derived by the aforementioned least squares program, were found to be $a = 13.029 \text{ \AA}$ and $c = 13.847 \text{ \AA}$.

The volume of the unit cells of the various leucites derived from the x-ray calculations are given below in Table 6 together with the densities derived from them. In computing the density the value of Z , the number of formula weights of KAlSi_2O_6 contained in the unit cell, is taken as 16 based on the work of WYART (1940). Chemical data for K_2O , Na_2O ,

Table 5. *X-ray oscillating crystal data for leucite from Alban Hills, Italy.*
Based on the measurements of von NÁRAY-SZABÓ (1942)

[CuK α , $\lambda = 1.5418 \text{ \AA}$]

| hkl | d (Calc.) | d (Obs.) | I | hkl | d (Calc.) | d (Obs.) | I |
|------|-----------|----------|-----|------|-----------|----------|-----|
| 101 | 9.489 | 9.509 | θ | 620 | 2.060 | 2.061 | 5 |
| 200 | 6.515 | 6.530 | θ | 226* | 2.061 | 2.060 | 2 |
| 112 | 5.535 | 5.546 | 5 | 541 | 2.013 | 2.013 | 2 |
| 211 | 5.371 | 5.392 | 5 | 622 | 1.975 | 1.976 | 2 |
| 202 | 4.744 | 4.730 | 2 | 631 | 1.923 | 1.922 | 4 |
| 220 | 4.606 | 4.618 | 2 | 444* | 1.916 | 1.916 | 7-8 |
| 222 | 3.835 | 3.834 | θ | 534 | 1.877 | 1.875 | 2 |
| 213 | 3.618 | 3.619 | 4 | 217 | 1.873 | 1.871 | 5 |
| 312 | 3.541 | 3.537 | 4 | 543 | 1.862 | 1.862 | 3 |
| 321 | 3.496 | 3.504 | 4 | 604 | 1.840 | 1.840 | 5 |
| 004 | 3.462 | 3.456 | 16 | 426 | 1.809 | 1.810 | 3 |
| 400 | 3.257 | 3.253 | 20 | 640 | 1.807 | 1.805 | 4 |
| 114 | 3.241 | 3.239 | θ | 633 | 1.790 | 1.789 | 3 |
| 303* | 3.159 | 3.157 | 2 | 712 | 1.781 | 1.780 | 5 |
| 411 | 3.081 | 3.083 | 2 | 721 | 1.775 | 1.776 | 5 |
| 204 | 3.057 | 3.058 | 10 | 642 | 1.748 | 1.748 | 3 |
| 402 | 2.947 | 2.954 | 10 | 008 | 1.731 | 1.733 | 3 |
| 420 | 2.913 | 2.908 | 10 | 703 | 1.726 | 1.725 | 5 |
| 323* | 2.843 | 2.843 | 9 | 615 | 1.694 | 1.694 | 3 |
| 332 | 2.807 | 2.804 | 9 | 723 | 1.669 | 1.668 | 9 |
| 422 | 2.685 | 2.686 | 1 | 732 | 1.661 | 1.661 | 8 |
| 314 | 2.650 | 2.649 | 4 | 800 | 1.629 | 1.630 | 7 |
| 413 | 2.607 | 2.604 | 3 | 714 | 1.627 | 1.626 | 5 |
| 431 | 2.561 | 2.561 | 4 | 811* | 1.604 | 1.604 | 4 |
| 501* | 2.559 | 2.559 | 1-2 | 741 | 1.605 | 1.606 | 4 |
| 512 | 2.397 | 2.395 | 2 | 802 | 1.585 | 1.586 | 7 |
| 521 | 2.383 | 2.386 | 3 | 820 | 1.580 | 1.580 | 8 |
| 404 | 2.372 | 2.371 | 8 | 507 | 1.576 | 1.576 | 3 |
| 305 | 2.335 | 2.336 | 3 | 822 | 1.540 | 1.542 | 3 |
| 440 | 2.303 | 2.301 | 8 | 660 | 1.535 | 1.536 | 2 |
| 433 | 2.269 | 2.267 | 3 | 734 | 1.534 | 1.533 | 3 |
| 503* | 2.267 | 2.265 | 3 | 743 | 1.525 | 1.527 | 3 |
| 206 | 2.175 | 2.178 | 5 | 831 | 1.516 | 1.514 | 2-3 |
| 600 | 2.172 | 2.172 | θ | 725 | 1.503 | 1.503 | 4 |
| 523 | 2.143 | 2.141 | 3 | 752 | 1.480 | 1.480 | 4 |
| 532 | 2.126 | 2.123 | 3-4 | 804 | 1.474 | 1.474 | 6 |
| 611 | 2.117 | 2.117 | 3 | 833 | 1.448 | 1.449 | 2 |

* Omitted by mistake, values hand computed.
I = θ signifies that no data are available.

Table 6. *Volume of the unit cell and density for six natural leucites, a synthetic leucite and iron-leucite measured at 25° C*
 (Partial chemical data given for comparison)

| Locality or Origin | Volume of unit cell | Density | K ₂ O | Na ₂ O | CaO | Fe ₂ O ₃ | FeO |
|--|---------------------|---------|--------------------|-------------------|------|--------------------------------|------|
| Garofali, Roccamonfina, Italy | (A°) ³ | | | | | | |
| | 2352.8 | 2.464 | 19.8 | 0.95 | 0.14 | 0.29 | |
| Synthetic leucite | 2348.3 | 2.469 | "21.59" [theo.] | 0.10 | 0.04 | None | |
| Nyiragongo, The Congo | 2346.3 | 2.471 | 19.87 | 0.95 | 0.09 | 0.53 | 0.14 |
| Villa Senni, Alban Hills, Italy | 2345.5 | 2.472 | 21.1 | 0.28 | 0.12 | 0.43 | |
| Vesuvius, Italy | 2338.9 | 2.479 | 19.4 | 1.24 | 0.14 | 0.29 | |
| Pompeii, Italy | 2337.6 | 2.480 | 21.0 | 0.26 | 0.16 | 0.71 | |
| Monte Somma, Italy | 2334.6 | 2.484 | 20.0 | 1.04 | 0.14 | 0.29 | |
| Synthetic iron-leucite | 2435.8 | 2.695 | | | | | |
| VON NÁRAY SZABÓ's leucite from Vesuvius recalculated in this study | 2350.6 | 2.467 | | | | | |

CaO, Fe₂O₃ and FeO are also given in this table for comparison. No clear-cut correlation is apparent and because the quantities of these elements are small this may arise from a disparity between the chemical and spectrographic data. Although the number of structural formula based on chemical data in Table 9 are too few in number to draw a conclusion there may be a relationship between Al in tetrahedral coordination and density.

The inversion of leucite is accompanied by a volume change, Δv , which is of such proportion that its effect was easily observed under the microscope by BOWEN and SCHAIRER (1929) who write:

"At the inversion point, with falling temperature, leucite suffers a notable contraction of volume. In consequence of this each leucite crystal in our intermediate preparations, quenched from a high temperature, is surrounded by a halo of highly-strained, birefracting glass in which γ is radially disposed. The glass is in a state of radial tension. The leucite crystals themselves must be in a like condition."

Using the data of WYART (1940) for the unit cell of the cubic, high temperature, form of leucite with $a_0 = 13.43$ and his observation that Z is also 16, the volume and density of his synthetic leucite, which had

been synthesized by Charles and Georges Friedel, were calculated and found be 2422 (Å)³ and 2.39 respectively. For the tetragonal polymorph of this synthetic leucite the volume and density are 2305 and 2.50 respectively. From these data the calculated value of Δv is:

$$[\Delta v_{\text{cubic} \rightarrow \text{tet}} = -117.0 \text{ (Å)}^3]$$

or a diminution of 4.83 percent.

Thermal Analysis

Inversion Phenomena of Leucite as Disclosed by DTA Studies

The differential thermal analysis curves used in this study were obtained as photographs. The apparatus and technique have been described by FAUST (1948 and 1950). The samples were heated at the rate of 12° C per minute.

The DTA curves, figures 1, 2 and 3, show that the inversion of leucite is not a simple displacive transformation (nomenclature of BUERGER, 1951) such as characterizes quartz but rather is a compound transformation consisting of two separate thermodynamic processes. The first endothermic trough is broad and round; the second is narrow and pointed.

The DTA curves for pure synthetic leucite are given in figure 1. The magnitude of the effect for various sensitivities of the galvanometer assembly, curves C-717, C-718, C-719 and C-720, shows clearly that, for the apparatus used in this study, a resistance of 100 ohms in the differential galvanometer circuit gives the most useful curves. The measurement of the temperatures of maxima and minima becomes increasingly difficult in less sensitive arrangements.

For mixtures of synthetic leucite and synthetic diopside, curves C-691 and C-696, the results are not too encouraging since only a broad endothermic trough appears.

The temperature data for the inversion are given in Table 7. Curve C-720 was selected as the best record for the determination of the inversion temperatures. At 602° C the inversion phenomena begin and culminates in a major endothermic trough at 631° C. This is followed by an inflection in the curve at 648° and the inversion process is completed at 656° C. Unfortunately, the differential thermal curve for the cooling process was not recorded at this time. Record C-764 (not shown in the figure) obtained on 0.3967 gms of sample shows the differential thermal

curve for both the heating and cooling process. While the maxima and minima are not recorded as sharply as in curve C-720, since there was considerably less sample available, the curves show clearly that the inversion of leucite is a reversible phenomenon, and that it is *not a simple isothermal process*.

The DTA curves for natural leucites are given in figures 2 and 3, and the data are summarized in Table 8. The DTA curves for natural leucite are much more manifest in character than those for the synthetic leucite. A comparison of the DTA records shows that curve C-596 approaches more closely the form of the curve for synthetic

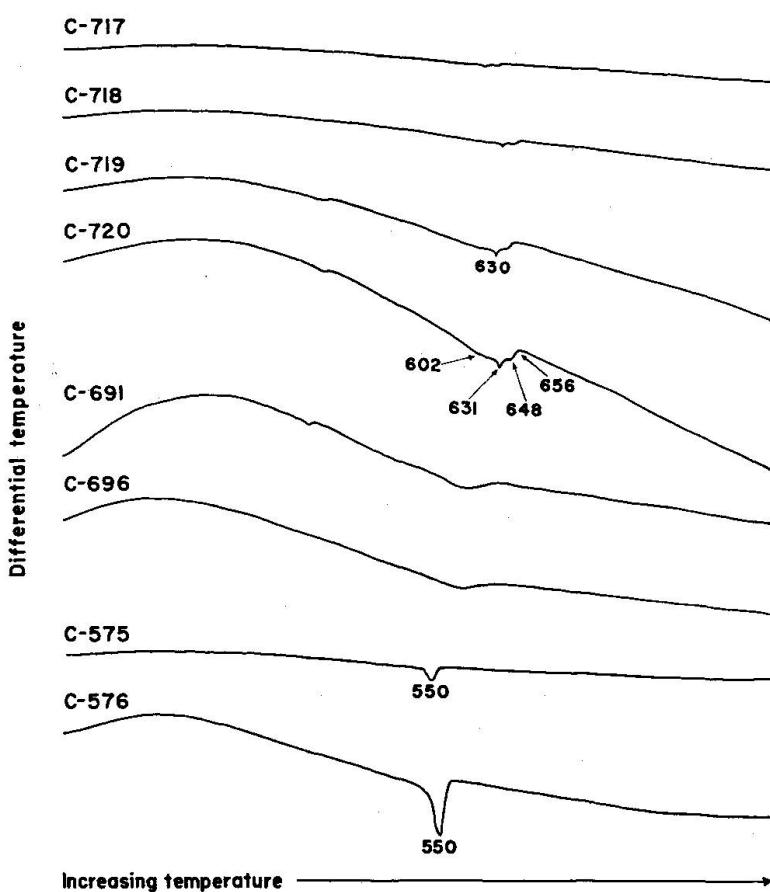


Fig. 1. Differential thermal analysis curves of synthetic leucites and iron-leucite. C-717, synthetic leucite, 600 ohms resistance in galvanometer circuit, 0.5082 grams; C-718 a re-run of C-717 at 300 ohms; C-719 a re-run of C-718 at 100 ohms; C-720 a re-run of C-719 at 50 ohms. C-691, mixture of 95 per cent synthetic leucite and 5 per cent synthetic diopside, at 100 ohms, 0.4075 grams; C-696 mixture of 80 per cent synthetic leucite and 20 per cent synthetic diopside, at 100 ohms, 0.5675 grams. C-575, synthetic iron-leucite, at 600 ohms, 0.4824 grams; C-576 a re-run of C-575 at 100 ohms.

leucite. The inversion internal for the synthetic material appears to be longer than that for the natural leucites and this will reduce the sharpness of the troughs and peaks. The explanation for this difference in character is not now known but it may be related to the state of subdivision of the materials and the possible presence of a thin selvage of glass around the synthetic crystals.

The DTA curves and the data in Tables 7 and 8 show clearly that the inversion phenomena in natural leucites is a reversible reaction. The agreement between the data for the heating curves for leucites No. 5 and 7, presumably from the same lava, is very good, the greatest dif-

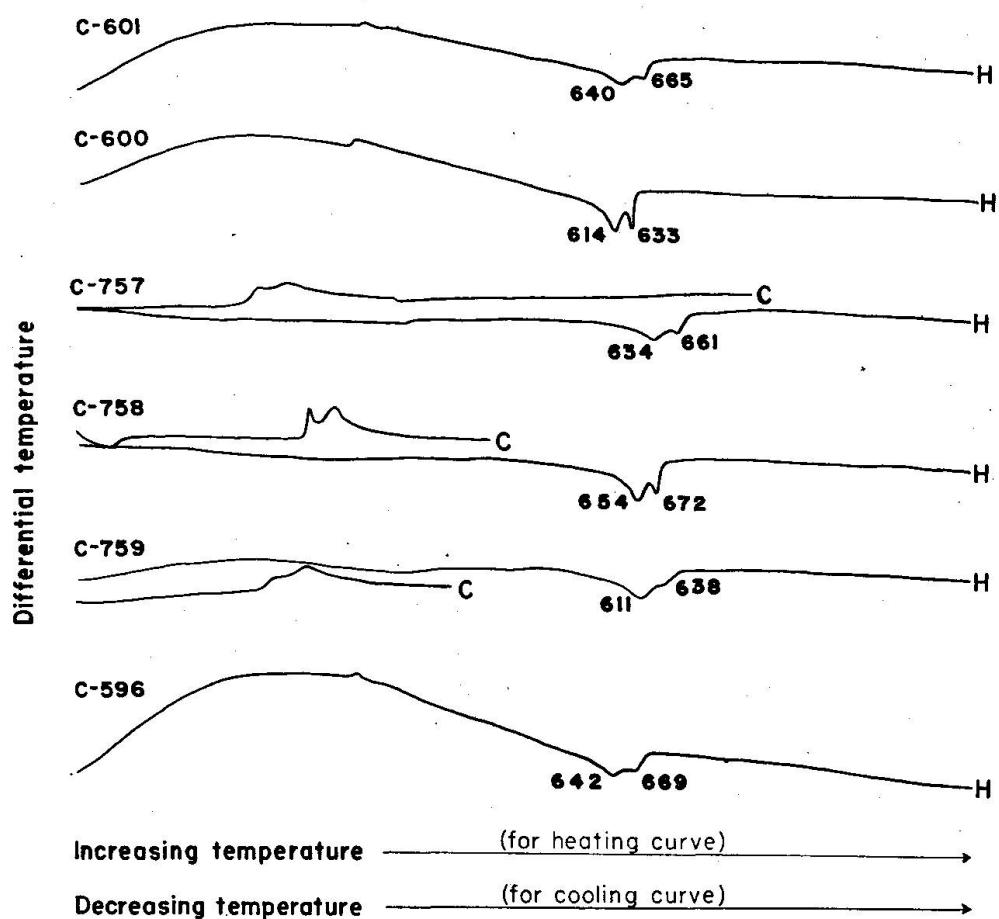


Fig. 2. Differential thermal analysis curves of natural leucites from various Italian localities. C-601, leucite No. 5, 0.4830 grams; C-600, leucite No. 6, 0.42 ± grams; C-757, leucite No. 7, 0.4312 grams; C-758, leucite No. 8, 0.4759 grams; C-759, leucite No. 9, 0.5092 grams; C-596, leucite No. 10, 0.3104 grams. All temperatures given in degrees Centigrade. These curves were all obtained with a resistance of 100 ohms in the galvanometer circuit. Curve marked "H" at the termination is a heating curve, curve marked "C" is a cooling curve.

Table 7. Observations on the thermal analysis curves of synthetic leucite and iron-leucite

| Synthetic Preparation | Record No. | Heating Curve | | | | Resistance in galvanometer circuit (ohms) | Weight of sample used (gram) |
|-----------------------|------------|---------------|-----------------------------|--------------|-------------------------|---|------------------------------|
| | | Inflexion °C | Major endothermic trough °C | Inflexion °C | End of the inflexion °C | | |
| Leucite No. 1 | C-719 | Present | 632 | Present | 656 | 100 | 0.5082 |
| Leucite No. 1 | C-720 | 602 | 631 | 648 | A | 50 | 0.5082 |
| Leucite No. 1 | C-764 | Present | 624 | Present | A | 100 | 0.3967 |
| Leucite No. 1 | C-766 | Present | 618 | Present | B | 100 | 0.3 ± |
| Iron-leucite | C-575 | — | 550 | — | — | 600 | 0.4824 |
| Iron-leucite | C-576 | — | 550 | — | — | 100 | 0.4824 |

Table 8. Observations on the thermal analysis curves of natural leucite

| Leucite No. | Record No. | Min. of broad endothermic trough, °C | Minimum of sharp trough, °C | Interval between minima, °C | Resistance in galvanometer circuit (ohms) | Weight of sample used (gram) | Notes |
|-------------|------------|--------------------------------------|-----------------------------|-----------------------------|---|------------------------------|-----------------|
| 5 | C-601 | 640 ¹⁾ | 665 ¹⁾ | 25 | 100 | 0.4830 | 1) BERKEL- |
| 6 | C-600 | 614 | 633 | 19 | 100 | 0.42 ± | HAMER (1944); |
| 7 | C-757 | 634 | 661 | 27 | 100 | 0.4312 | est. as 650°C ± |
| 9 | C-759 | 611 | 638 | 27 | 100 | 0.5092 | and 675°C ± by |
| 10 | C-596 | 642 | 669 | 27 | 100 | 0.3104 | G.T.F. |
| T.G.S.-1 | C-781 | 637 | 656 | 19 | 100 | 0.4422 | |
| T.G.S.-2 | C-779 | 637 | 664 | 27 | 100 | 0.4525 | |
| T.G.S.-3 | C-783 | 637 | 674 | 37 | 50 | 0.4820 | |

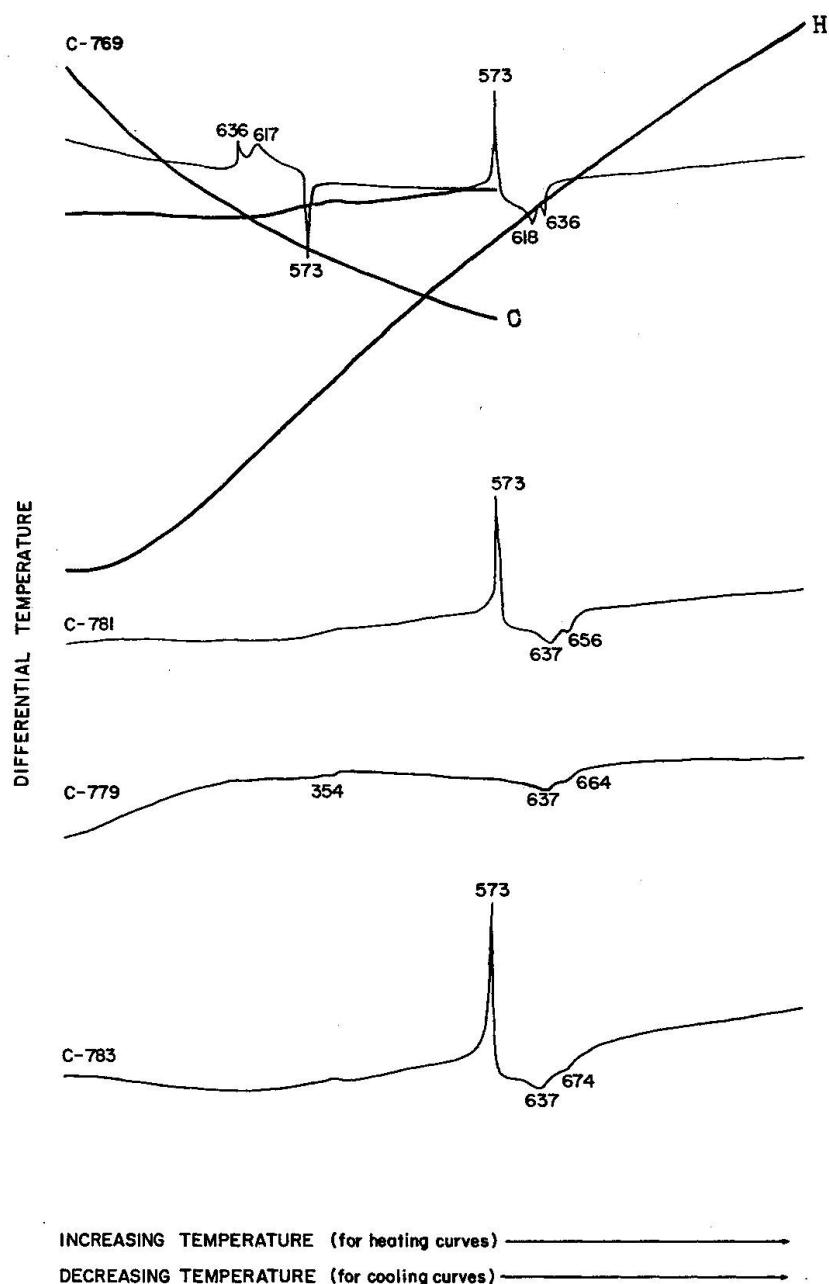


Fig. 3. Differential thermal analysis curves of leucites. C-769, leucite No. 8 obtained with quartz as the neutral body. Heating curve H and cooling curve C. Weight of leucite No. 8 used is 0.4548 grams that of quartz 0.5146 grams. C-781, leucite No. T.G.S.-1, 0.4422 grams; C-779, leucite No. T.G.S.-2, 0.4525 grams; C-783, leucite No. T.G.S.-3, 0.4820 grams. Curves C-781 and C-783 were run against quartz as the neutral body. Resistance in series with the galvanometer circuit 100 ohms, except for C-783 for which the value was 50 ohms.

ference 6° C is associated with the broad trough, for which a minimum is more difficult to assign.

To confirm the isothermal character of the transformations associated with the broad endothermic trough and the sharp endothermic trough a sample of natural leucite from Vesuvius, Italy (Leucite No. 8) was run against quartz as the neutral body. The low-high and high-low inversions of quartz then served as a calibration for the heating and cooling cycles respectively. The record of this curve, C-769, is given as figure 3. The minimum of the broad round endotherm is at 618° C while the corresponding maximum of the exotherm is at 617° C. The minimum of the sharp endotherm is at 636° C and the corresponding maximum of the exotherm is at 636° C. These data show no hysteresis associated with the inversion phenomena.

The *compound character* of the inversion of leucite, shown by the presence of two endotherms, does not arise from solid solution relationships involving Na_2O as suggested by RINNE and KOLB (1910) for it is present in pure leucite. The phase-equilibrium study of the system $\text{K}_2\text{O}-\text{Na}_2\text{O}-\text{Al}_2\text{O}_3-\text{SiO}_2$ by SCHAIRER and BOWEN (1947) shows that leucites cannot contain more than 1.0 to 1.5 per cent of Na_2O . Recent analysis of leucites fall within this range and show only small amounts of Na_2O , less than one per cent. The analysis presented in Table 2 confirm the limited content of both Na_2O and CaO in leucite. It thus appears that the compound character of the inversion is associated with the thermal stability of the crystal structures of the polymorphs.

The two separate endotherms represent two distinct thermodynamic processes. These endotherms may be interpreted in one of several ways, thus: (1) there may be three phases involved in their origin — a low-temperature tetragonal phase giving rise to the first trough in its inversion to an intermediate phase, the intermediate phase in its turn inverting to the high-temperature cubic phase, and giving rise to the second trough in this inversion; or (2) the partial coalescence of the troughs in some of the curves may be an indication of the nonexistence of an intermediate phase; and this anomalous behavior may be attributed to a sluggishness in its transformation from the tetragonal to the cubic form. This is a phenomenon involving order and disorder of atoms in the crystal structure. BRAGG and WILLIAMS (1934) in a study of alloys showed that, when atoms pass from disordered to ordered positions, there is an evolution of energy, expressed as a loss of heat, and reciprocally an absorption of heat on passing from ordered to disordered positions.

In leucite, which contains Al^{3+} and Si^{4+} ions, it may be expected that Al - Si order-disorder relations would obtain because in the feldspars, microcline and sanidine which are polymorphs, GOLDSMITH and LAVES (1954) have given evidence in support of such Al - Si order-disorder.

The origin of the compound character of the leucite inversion is not explained in this paper, it is under investigation by means of a high-temperature x-ray camera and will be reported on later.

Inversion Phenomena of Low Iron-leucite to High Iron-leucite

(1) Cooling curve study

A sample of well crystallized iron-leucite was placed in a platinum crucible. A platinum: platinum 90%:rhodium, 10%:thermocouple was inserted into the iron-leucite and the whole assembly lowered by means of heavy platinum wire into the "hot spot" of a "Geophysical Laboratory type" quenching furnace (FAUST, 1936). A number of heating and cooling curves were made. Four of the cooling curves included the range of temperatures over which the transformation high iron-leucite to low iron-leucite takes place. Graphical analysis of the data fails to show a significant break in the cooling curve. Inspection of the temperature measurements with a knowledge of the DTA data shows that the temperature range over which the transformation occurred almost coincided with the interval over which a change in rate of the furnace cooling took place. This overlap could easily mask any decisive break in the cooling curve (see fig. 1).

(2) DTA Studies

Differential thermal analysis of iron-leucite shows a single endothermic trough, with the temperature of the minimum at $550^{\circ} C$. The trough while steep and narrow is definitely not as steep nor as pointed as that shown by quartz under similar experimental conditions (see fig. 1, curve C-575).

The single thermodynamic process involved in the inversion of low iron-leucite to high iron-leucite is in marked discord with the behavior of its aluminum analogue. The failure of iron-leucite to show a double set of troughs, in the inversion process, may arise from the volume relations of the two ions Al^{3+} and Fe^{3+} which, according to Pauling, have ionic radii of 0.50 \AA and 0.64 \AA respectively (Fe^{3+} is 12.8 percent larger). The size of the Fe^{3+} with respect to the Si^{4+} ion, whose ionic

radius is 0.41 Å, is such that no Fe^{3+} – Si^{4+} order-disorder can easily take place and accordingly iron-leucite is an ordered structure. Based on this reasoning, it might be concluded that the second hypothesis (that of Al–Si order-disorder) is the cause of the compound character of the inversion of natural leucites. Further details on this will be published later.

It is of interest to note that COOMBS (1954) in a study of the naturally occurring iron-orthoclase from Madagascar suggested that a high Fe^{3+} concentration distributed in the $[\text{AlSi}_3]$ positions, in the tetrahedral coordination, tended to inhibit the ordering process. WONES and APPLEMEN (1962) show evidence that iron-microcline is more stable than iron-sanidine at temperatures up to $704^\circ \pm 6^\circ \text{ C}$ at which point a first order transition takes place. Hence if iron-microcline is ordered, the Fe^{3+} would promote ordered structures at higher temperatures.

Petrogenetic Significance of the Inversion of Leucite

BERNHARD VON COTTA (1858) in a book entitled „Geologische Fragen“ made the following significant statement:

„Kein Gestein verharrt vollkommen in dem Zustande seiner ersten Entstehung.“

Almost a hundred years later Professor TOMKEIEFF (1954) in his recent book has refocussed attention on VON COTTA's observation translating it to “No rock remains completely in the state of its original formation.”

This is a clear-cut statement of the problem which has occupied the foremost attention of petrologists and mineralogists in recent years, namely, the characteristics and properties of the pyrogenic minerals, (ZIRKEL, 1866) those that crystallize directly from the magma. TUTTLE (1952) has made a notable contribution in the study of quartz and the feldspars. BOWEN and SCHAIRER, and coworkers (1930, 1933, 1935), TOMITA (1934), HESS (1941, 1949) and others have made similar studies of the pyroxenes.

In a cooling magma, which represents a natural quench, there is a constant adjustment between the crystal phases and the melt as the temperature drops. For solid phases there is the adjustment due to contraction on cooling and that due to the ensuing jostling of the grains when the mass becomes a rigid framework. Superimposed upon these there may be inversion phenomena, reaction of incongruently melting phases with the melt, recrystallization phenomena, unmixing of solid

solutions, diffusion of gases to form vesicles, etc. The most significant advance in phase rule studies of the silicates in the last 25 years has been the realization of the importance of sub-liquidus phenomena. With a thorough correlation of the sub-liquidus phenomena, the mineralogical and petrological observations and the field relationships we shall be able to interpret the history of igneous rocks. Leucite represents a pyrogenic mineral that undergoes sub-liquidus changes.

BOWEN and SCHAIRER (1929) pointed out the importance of the leucite inversion in petrology. First, the inversion temperature of leucite is "a good indication of the minimum temperature of crystallization of leucite and second that „the inversion of leucite is a fundamental control in the development of an important constituent of many alkaline rocks, viz. pseudo-leucite".

In order to assess the significances of the phase leucite as a geological thermometer it is necessary to consider its morphology, crystal size, growth, composition, and environment during crystallization.

The simplicity of the morphology of leucite is striking. It crystallizes at high temperatures as well formed tetragonal trisoctahedrons {211} (sometimes called the leucitohedron). Rarely do the cube {100} and the rhombic dodecahedron {110} occur and then usually as subordinate forms (GOLDSCHMIDT, 1918). Upon cooling down to earth-surface temperatures it transforms to a tetragonal symmetry, a pseudomorphous form, and this is accompanied by a twinning parallel to the rhombic dodecahedron {110}. The twinning process gives rise to striations on the crystal faces. These striated surfaces are not coplanar, but form re-entrant angles. ROSENBUSCH (1885) studied the surfaces of a twinned crystal of leucite from Vesuvius in reflected light and at high temperature and observed that the re-entrant angles formed by the twinning lamellae disappeared into the tetragonal trisoctahedral plane {211} at the inversion point. Upon cooling the twinning lamellae on {110} re-appear but not necessarily in their previous positions on the crystal faces.

All natural crystals so far found are pseudomorphs and they are always twinned — the so called mimetic twins which simulate to a higher symmetry (MÜGGE, 1901).

The size of leucite crystals, as reported in the literature, varies from the tiny crystals in the groundmass of rocks, about .01 to .03 mm, to coarse crystals attaining 5 to 6 centimeters in their longest dimension. SINIGALLIA (1891, p. 418) described crystals of leucite in a glassy rock from the 1753 lava of Vesuvius as follows:

„Der Leucit bildet zum Teil grössere Krystalle, welche sich oft als Haufwerk kleinerer herausstellen. Ihre Formen sind im Allgemeinen um so schärfster und um so mehr im Gleichgewicht entwickelt, je kleiner die Individuen sind.“

To gain some idea of the size and range in size of leucite crystals at a given locality the leucite crystals in the collection of the U. S. National Museum were measured parallel to a pseudocubic axis with the following results: — (a) Vesuvius, Campania Region, Italy, two large crystals of 6.0 and 4.0 cm respectively and 34 crystals ranging from 2.7 to 0.4 cm; Monte Somma, Vesuvius, 2 crystals of 2 cm each; Roccamonfina, Campania Region, Italy, 11 crystals in the size range of 4.5 to 3.0 cm, and 5 crystals in the range 2.6 to 1.6 cm; Picomontino, near Naples, Italy, a single crystal of 2.0 cm; Villa Senni, Alban Hills, Italy, many small crystals in the range of 0.8 to 0.3 cm (italite); Petrisco, above San Martino, Viterbo, Italy, many small crystals in the range 0.9 to 0.5 cm; Rocca Vulcano, Italy, 4 crystals in the range of 2.3 to 1.7 cm; Magnet Cove, Arkansas, 10 crystals ranging from 4.5 to 2.8 cm, and 4 crystals ranging from 2.3 to 1.6 cm; Serra dos Pocos de Caldas, SW Minas Geraes, Brazil, 4 crystals in the size range of 4.5 to 3.2 cm, and 5 in the range of 2.0 to 0.8 cm.

These measurements confirm the general statements found in the mineralogical literature. Moreover, they show that in some rocks, as the italite, the span of crystal sizes may be gradational. PALACHE (1932) has emphasized the need of collecting data on crystal size.

Knowledge of the growth of leucite crystals was inferred from the examination of thin sections of natural crystals and from studies of synthetic leucites grown in melts. The skeletal growth of leucite was noticed early by petrographers and ZIRKEL (1893) has discussed this in detail. In many crystals of leucite, as viewed in the octagonal cross section, an arrangement of tiny prisms parallel to the exterior octagonal cross sections was observed. These prisms are commonly greenish or yellowish augite and colorless microlites of feldspars. Rounded, club-shaped, and oval-shaped, black and brownish translucent grains of glassy or slaggy material are frequently found and the glass usually crystallizes and magnetite separates out. Gas cavities have also been described. Particularly informative observations on the skeletal growth of leucite were made by WEED and PIRSSON (1896, p. 144—146) who found that the leucites of the groundmass showed a definite order of growth. This order is — (1) a cruciform growth along the cubic axes; (2) the ends of the cross develop a club-shape; (3) these ends grow by extension along the boundaries of the tetragonal trisoctahedron, and glass is

enclosed within the skeletal form; (4) starting from the center of the cross, growth begins along the dodecahedral axes; (5) the growth started in (4) advances to close the polygon and form a wheel-shaped figure. The final stage consists of the growth of material along the octahedral axes and the completion of the outer surface of the tetragonal trisoctahedron and when the process of growth ceases and the crystal then has club-shaped inclusions of glass projecting inward toward the center.

The growth of leucite in mineral synthesis studies was described by FOUQUÉ and MICHEL-LÉVY (1882) who noted that:

"The crystallites of leucite appear as dendrites, sensibly rectangular, of which each unit is a small crystal generally with sharp forms and appearing in profile with the faces (211), (100), (110). The junctions of these crystals are along the faces (110) and their aggregation constitutes a grouping of six units in a right angle, in such a way that, in section, one often sees a regular cross."

They also observed the formation of a rock, leucite-tephrite, made up of 60 percent leucite, 31 percent of labradorite and 9 percent of augite in which the labradorite and augite formed late and were squeezed into the interstices of the leucite crystal framework. The leucite crystals were 0.35 mm in section, the labradorite laths — 0.02 to 0.05 mm long and the augite prisms 0.025—0.005 mm long. NEUBAUER (1911) similarly synthesized leucite-bearing rocks and noticed that leucite crystallized easily from the liquid phase.

Leucite crystals formed in the phase rule studies of BOWEN and SCHAIKER (1929) "are usually of a rounded outline but sometimes are well developed icositetrahedra." FAUST (1936) observed that "iron-leucite crystals developed as subhedral grains which have a strong tendency to arrange themselves in star-like or ray-like patterns," that is skeletal growths.

Since the growth of crystals from melts is a rate-process the calculation of the absolute rates of growth applicable to all magmas is not possible. Consequently such experimental data as are available must be judged in terms of the physicochemical conditions of the systems studied. DOELTER (1903), for example, measured the rate of growth of leucite crystals from a melt and then calculated that: "the larger crystals of leucite which are often 2 cm thick, could have grown from the lava, at atmospheric pressure, in about 800 to 1600 hours". This calculation was based on the assumption that the temperature during the period of crystallization did not drop more than 10° C, for Doelter found that constancy of temperature is a condition for the growth of large crystals.

The velocity of crystallization of leucite from artificial silicate has

also been studied by KITTL (1912) a student of Doelter. He melted a crystal of leucite, from Vesuvius, in a „Doelter Heizmikroskop“ and them observed the growth of the granular crystals in the melt. In cooling from a temperature of 1330° to 1320° in 5 minutes, leucite crystals grew 0.03 mm; from 1320° to 1310° in a time interval of five minutes they grew to 0.06—0.09 min., and in the interval of 1310 to 1300° C their size did not change.

The composition of natural leucites, as recorded in the standard reference works, shows (1) that the calcium oxide content is rather low, with relatively few exceptions, less than one percent and recent analyses (see Table 2) generally show between 0.10 and 0.20 percent; (2) sodium oxide content is generally less than two percent, and its value is possibly related to the petrographic province from which the leucites come; (3) ferric oxide, when determined, ranges from about 0.15 to a little over one percent; (4) ferrous oxide is still lower in amounts generally up to about 0.25 percent. The analyses reported in this study together with an earlier analysis by STEIGER (in CLARKE and STEIGER, 1900), made in the U. S. Geological Survey, were calculated in terms of the structural formula for $1/16$ the unit cell content and are given in Table 9.

Table 9. *Structural formulas for leucites studied in this research and an average leucite from Vesuvius, Italy*

| | |
|---|--|
| Nyiragongo, The Congo | 16 ([K _{.92} Na _{.07} Ca _{.01}] [Fe _{.02} ³⁺ Al _{.98}] [Si _{1.99} Al _{.01}]O ₆) |
| Mt. Mikeno, The Congo ¹⁾ | 16 ([K _{.98} Na _{.05} Ca _{.01}] [Fe _{.01} ³⁺ Fe _{.01} ²⁺ Al _{.97}] [Si _{1.99} Al _{.01}]O ₆) |
| Villa Senni, Italy ²⁾ | 16 ([K _{1.01} Na _{.02} Ca _{.01}] [Fe _{.03} ³⁺ Al _{.95}] [Si _{2.01}]O ₆) |
| Vesuvius, Italy (STEIGER's analysis) | 16 ([K _{.90} Na _{.09} Ca _{.01}] [Al _{1.01}] [Si _{2.00}]O ₆) |
| Vesuvius, Italy (TSCHIR- WINSKY's Average of 14 leucites) | 16 ([K _{.82} Na _{.15} Ca _{.01}] [Al _{.98}] [Si _{2.02}]O ₆) |

¹⁾ The Ca is given as .01 it is actually .003.

²⁾ The Ca is given as .01, it is actually .005.

All of the analyses of natural crystals of leucite show only a small amount of Fe₂O₃ indicating that the quantity of iron-leucite in solid solution with leucite is small.

TSCHIRWINSKY (1931) averaged fourteen analyses of leucite from Vesuvius, and SAHAMA (1952) calculated from this average the ratio of Na:K to be 15.4 to 84.6 percent. He suggested that this appears to be a characteristic of the petrographic province of Vesuvius.

The relatively low amounts of sodium in leucites is, however, noteworthy. SHAND (1943, p. 27) has observed that despite the experiments of LEMBERG (1876), who showed that leucite ($KAlSi_2O_6$) may be treated with a solution of sodium chloride at temperatures of 180—195° C and converted to analcime ($NaAlSi_2O_6 \cdot H_2O$) and conversely analcime reacted with potassium chloride to form leucite, that natural leucites contain very little soda. As SCHAIRER and BOWEN (1947) have shown no such interchange of cations exists at the liquidus temperature in anhydrous silicate melts. FUDALI (1957, 1961) has supplied the answer for some of these problems. He studied the system nepheline-kalsilite-silica at $p_{H_2O} = 1000 \text{ kg/cm}^2$ and found that liquidus temperatures are "substantially lower than in the dry system and the primary field of leucite is considerably reduced." Further, leucite solid solutions containing a maximum of 40 weight percent of $NaAlSi_2O_6$ were prepared. These "leucite solid solutions become unstable at high temperatures as the sodium content increases." In extrusive rocks "the subsolidus breakdown is responsible for the formation of pseudoleucite" and in "most plutonic alkaline rocks whose compositions lie in the primary field of leucite and which consist essentially of nepheline and sanidine attain this final crystalline assemblage through the subsolidus breakdown of soda-rich leucite."

The environment in which leucite crystals grew during their crystallization may be inferred from the preceding data and observations. The large size of the phenocrysts suggests that they grew rapidly in a fluid rather than a visous melt. Their skeletal habit suggests rapid growth. The inclusions of other crystal phases within the leucites is generally assumed to indicate that the included phases grew first, but it could also be interpreted as a more rapid rate of growth of the leucite which then overtook its associates and enclosed them. The inclusion of globules of liquid lava entrapped within the dendritic forms suggests that a quenching of the crystallizing system took place rather abruptly as in a volcanic eruption — when ejectamenta is cast out of a vent or lava is extruded. In many leucitic rocks, leucite occurs in both phenocrysts and groundmass which would necessitate a rather long period of crystallization.

Dendritic growth of leucite crystals, described by several scientists, is generally associated with supercooling. The well known process of producing dendritic crystals by pouring the liquid away from a partially crystallized ingot of metal, HUME-ROTHERY and RAYNOR (1954), may have a parallel in the crystallization of leucitic rocks. If a potassium-

rich differentiate, at a temperature well above the inversion point of leucite, were suddenly moved from a lower reservoir to a higher reservoir in the earth's surface, by a tectonic process, supercooling of the liquid mass would take place and dendritic crystals of leucite could form. Upon extrusion such dendritic crystals could continue to grow until natural quenching stopped crystallization.

Leucite is frequently the first phase to form in leucite-rich magmas. NEUBAUER (1911, p. 199) found in his "experiment II" that for a melt composed of 60 percent leucite, 25 percent augite and 15 percent orthoclase, gravitational differentiation took place. The upper portion of the melt was enriched in leucite crystals (many large ones) and the bottom portion of the melt was impoverished in leucite and contained only diopside crystals. LOEWINSON-LESSING (1899) had earlier studied the flotation of leucite crystals in Vesuvius lava and concluded that the leucite crystals arose by gravitational force. His experimental evidence was in concurrence with field observations in which it was noted that the upper layers of congealed lava were enriched in leucite crystals whereas the deeper layers in the section were richer in augite phenocrysts. TROMMSDORFF (1934) considered the problems of sinking and floating crystals hydrodynamically, and then examined the petrofabrics of rocks supposed to have formed by gravitational processes. For a leucite basanite from Vesuvius he determined the direction of flotation of the leucite crystals and sinking of the pyroxenes.

Leucite occurs almost exclusively in extrusive rocks and in the products of volcanic eruptions-lapilli, ejectamenta and tuffs. Its occurrence has been reported very rarely in metamorphic rocks. In rocks in which leucite is one of the primary phases its abundance ranges from traces up to ninety-four volume percent. These values represent extremes in differentiation products, the lowest value being present in some differentiates rich in nepheline, the highest in the rock italite described by WASHINGTON (1927). A study of the modes of leucite-bearing rocks in JOHANNSEN (1938, vol. 4) shows some very interesting, and not generally appreciated, data on the abundance of the phase leucite, at the time of its crystallization, in alkalic rocks. In summary the abundance of leucite in volume percent in the modes of these rocks is — leucite phonolite 8—14.4, leucite porphyry 27—40, leucite trachyte 8.8—40.6, campanite 30, sommaite 10—21, martinite 15.8—29.0, ottajanite 22—28, kivite 17, leucite shonkinite 20, orendite 30—40, jumillite 41, arkite 26—36.9, cocite 19, tavolite 37—37.1, vicoite 32.5—44.2, leucite tephrite-basanite 32.3—35.6, fiasconite 17.3—30, italite 90—93.5, fergusonite

40—49, missourite 16—28, wyomingite 26.1—35.7, vesbite 60, euktolith 30, katungite 10, batukite 12, madupite 20.3, kajanite 22 and schorensbergite 40. Similar and additional data are given by SHAND (1943) and TRÖGER (1935). The important point to be noted in this listing of the modal percentages of leucite is the significant abundance of leucite in these rocks. Thus, any change in the heat content, or the volume of the phase leucite at the inversion point, may affect the whole subsequent petrogenetic history of the rock. Moreover, the modal data for italite of 90—93.5 percent crystals of leucite makes possible an understanding of the "leucite rain" in the eruption of Vesuvius on April 22, 1845, mentioned by BLUM (1874, p. 308), when large quantities of leucite crystals were thrown out of the volcano.

In summary, the petrogenetic significance of the leucite inversion depends upon the caprice of nature. If a leucite-rich crystal-liquid differentiate is translated vertically by a tectonic process and is then, or later, extruded or ejected from a volcano the leucite crystals may persist because of the natural quenching of the lava. If, however, reaction can take place between the crystals and the lava or if the soda-rich leucite of FUDALI (1961) forms, the leucite will be replaced by a mixture composed of potassium feldspar and nepheline, pseudoleucite. If the leucite crystals persist in the congealed lava and are then percolated by sodium-rich groundwaters, Lembergs reaction of cation-exchange will obtain and the leucite will be converted into analcime.

Leucite has a strong tendency to crystallize and to form relatively coarse crystals. Some leucite-rich rocks approach a pegmatitic facies in grain size. The large crystal size and the polysynthetic twinning with its re-entrant angles present a large surface area for subsequent attack by solutions.

The experimental work in this paper shows that the inversion phenomena of leucite on cooling is a very complex one which instead of taking place as a prompt inversion such as the high to low inversion of quartz may actually span a temperature range of approximately 20°—30° C. This long span of temperature during which the atoms are undergoing changes in position as one crystal structure changes to another may play a part in the complex reaction forming pseudoleucite by creating a defect structure of lower stability than an annealed one. The volume change on inversion, a decrease of almost 5 percent, is accompanied by twinning with the formation of re-entrant angles on the crystal surfaces and by the establishment of strained conditions within the crystals and in the matrix.

The mass of rock affected by the volume change when leucite crystals make up 10 to 90 percent of the bulk is significant. For a rock containing 30—35 percent leucite, an average figure for the survey of modal data presented earlier in this paper, an overall decrease in volume of 1.5 percent would certainly increase the porosity of the rock and make the rock more susceptible to alteration.

That leucite is an excellent geological thermometer is attested to be the rapid reversibility of its transformation from cubic to tetragonal form and vice versa. The appearance of leucite paramorphs in the form of tetragonal trisoctahedrons proves that the leucite crystallized from the magma at temperatures above the inversion temperature.

The inversion temperature of any given sample of leucite can be readily measured by a calibrated DTA assembly, by an x-ray heating camera or by a microscope with a heating stage for the observation of birefringence.

The synthesis of apparently unrelated observations by others and the results of this study yields some interesting interpretations of leucite and its behavior in and out of the magma chamber. Thus, we may note that leucite crystallizes early and continues until the extruded lava solidifies as rock; that it accumulates by gravitational differentiation; and that quenching may take place so fast in volcanic ejectamenta that glass is retained as an inclusion in leucite crystals. The mere existence of leucite crystals in a lava indicates that lava was once at a temperature in excess of the inversion temperature.

In the course of the DTA studies several paramorphs of leucite, the so-called pseudoleucite, were examined. The DTA patterns that these samples yielded bear no resemblance to those of leucite.

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