

THE SYSTEM FORSTERITE-DIOPSIDE-AKERMANITE-  
LEUCITE AND ITS SIGNIFICANCE IN THE  
ORIGIN OF POTASSIUM-RICH MAFIC  
AND ULTRAMAFIC VOLCANIC ROCKS

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ABSTRACT

The phase relations of forsterite (Fo), diopside (Di), akermanite (Ak), and leucite (Lc), important end members of naturally-occurring potassium-rich mafic and ultramafic volcanic rocks, were determined experimentally at atmospheric pressure.

The following three joins were determined in the study: (1) diopside-akermanite-leucite, (2) forsterite-diopside-leucite, and (3) forsterite-akermanite-leucite. The phase assemblages, compositions, and temperatures of the piercing points in these joins are summarized below:

*Join*

|                                       |                          |                               |
|---------------------------------------|--------------------------|-------------------------------|
| (1) $Di_{ss} + Ak_{ss} + Lc_{ss} + L$ | $Di_{13}Ak_{29}Lc_{32}$  | $1281^{\circ} \pm 3^{\circ}C$ |
| (2) $Fo_{ss} + Di_{ss} + Lc_{ss} + L$ | $Fo_3Di_{60}Lc_{37}$     | $1296^{\circ} \pm 3^{\circ}C$ |
| (3) $Fo_{ss} + Mo_{ss} + Ak_{ss} + L$ | $Fo_{17}Ak_{78}Lc_5$     | $1423^{\circ} \pm 3^{\circ}C$ |
| (3) $Fo_{ss} + Ak_{ss} + Lc_{ss} + L$ | $Fo_9Ak_{43.5}Lc_{47.5}$ | $1286^{\circ} \pm 3^{\circ}C$ |

The join diopside-akermanite-leucite with 3 percent forsterite was also investigated to determine the temperature of the 5-phase assemblage, forsterite<sub>ss</sub> + diopside<sub>ss</sub> + akermanite<sub>ss</sub> + leucite<sub>ss</sub> + liquid. The fifth phase (forsterite<sub>ss</sub>) in this assemblage appears at  $1274^{\circ} \pm 4^{\circ}C$  and the solidus temperature occurs at  $1255^{\circ}C$ .

Study of the system suggests that an olivine-melilite leucitite magma can be generated from the following magma types: katungite, melilite leucitite, olivine melilitite, or olivine leucitite. A katungite magma of composition lying within the tetrahedron forsterite-diopside-akermanite-leucite can be derived from a monticellite alnoite magma because of a reaction relationship of monticellite with liquid.

INTRODUCTION

The potassium-rich mafic and ultramafic rocks have a distinct chemistry and mineralogy. They are characterized chemically by low  $SiO_2$ , low  $SiO_2/K_2O$  ratio, predominance of  $K_2O$  over  $Na_2O$  (Washington, 1906, pp. 183-186), and high  $TiO_2$  (Borley, 1967; Bell and Powell, 1969). These rocks also contain important, if minor, concentration of such elements as Ba, Rb, Sr, Zr, Nb, La, and Y (Higazy, 1954; Bell and Powell, 1969). The  $SiO_2$  content of the potassium-rich mafic and ultramafic rocks are usually less than 57 percent (Washington, 1906), but in some cases it may be less than 40 percent (Uganda, Holmes,

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1937; p. 207). The  $K_2O$  content of these rocks are usually above 2 percent (Table 14, Holmes, 1937; Tables 20 and 21, Turner and Verhoogen, 1960; Table 1, Washington, 1927), but in certain areas it may be higher than 19 percent (Washington, 1927; p. 178). The  $K_2O/Na_2O$  ratio of the potassium-rich mafic and ultramafic rocks is usually above 1, and in West Kimberley (Wade and Prider, 1940; p. 75) it exceeds 15. High-potassium content of these rocks and their under-saturated character are reflected by the presence of such minerals as leucite with or without other feldspathoids, olivine, and melilite.

The origin of these rocks has long been a difficult problem. It is not known whether they constitute independent magma series, a fractional crystallization product from one of the more commonly recognized magma types, or the result of assimilation or resorption of early-formed crystals. Before a more detailed assessment of their origin can be made a better understanding of the mutual phase relations of the various rocks and their relation to other igneous rock series is necessary.

The four essential minerals of most potassium-rich mafic and ultramafic rocks are leucite, augite, melilite, and olivine. Feldspar, phlogopite, or biotite may be present in minor amounts. When plagioclase is present melilite is absent. The bulk of the composition of these rocks

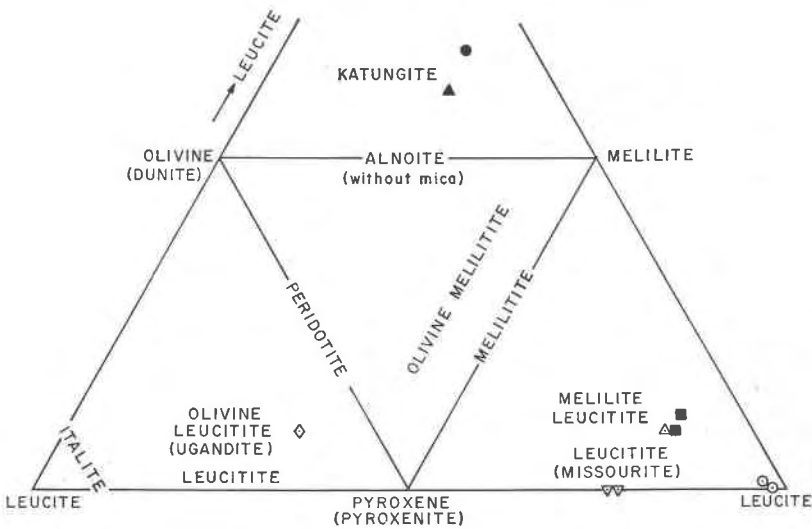


FIG. 1. Positions of different types of potassium-rich mafic and ultramafic volcanic rocks in the tetrahedron forsterite-diopside-akermanite-leucite.  $\odot$ —Italite,  $\nabla$ —Missourite (Washington, 1927),  $\diamond$ —Missourite (Weed and Pirsson, 1896),  $\triangle$ —Cecillite,  $\blacktriangle$ —Katungite,  $\bullet$ —Venanzite,  $\blacksquare$ —Vesbite.

thus lies within the phase volume of olivine-augite-melilite-leucite (Figure 1). A study of the simplified system forsterite-diopside-akermanite-leucite is thus applicable to the understanding of the phase relations and origin of potassium-rich mafic and ultramafic volcanic rocks.

#### PREVIOUS WORK

The system forsterite-diopside-akermanite and its related binary joins were studied by Ferguson and Merwin (1919). Bowen (1914) worked on the system diopside-forsterite-silica. Kushiro and Schairer (1963, 1964) reinvestigated the join diopside-forsterite and diopside-akermanite. Further investigations related to the system  $\text{CaO-MgO-SiO}_2$  were done by Bowen and others (1933), Osborn (1942, 1943), and Ricker and Osborn (1954). Bowen and Schairer (1929) worked on the system diopside-leucite. Data on the join leucite-forsterite are available from the system  $\text{K}_2\text{O-MgO-Al}_2\text{O}_3\text{-SiO}_2$  (Schairer, 1954).

#### EXPERIMENTAL METHOD

The same methods and techniques as described by Schairer (1959) were employed. Seventy-six mixtures were made to study the tetrahedron under consideration. Except for the mixture  $\text{Lc}_3\text{Ak}_{67}$ , all were homogenized to glasses and crystallized for a week at temperatures between  $800^\circ\text{-}1000^\circ\text{C}$ . The above-mentioned mixture was sintered at  $1400^\circ\text{C}$  for 2 hours, followed by crushing and reheating twice. The sintered material was then crystallized for a week at  $1000^\circ\text{C}$  and products are indicated on page 1247. Certified reagents of  $\text{CaCO}_3$ ,  $\text{MgO}$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{K}_2\text{CO}_3$ , and silica gel (Cat. No. ASTM D 1319-61T, S679, Grad 923) were used. The alkali content of the silica gel is less than 0.15 percent. Silica gel was heated at  $1000^\circ\text{C}$  to drive out the moisture. Potassium disilicate was prepared by the same method as described by Schairer and Bowen (1955). Homogeneity of the glasses was checked by measuring refractive indices. Platinum-wound resistance furnances were used for quenching. Quenching experiments were done in platinum envelopes (6 mm by 8 mm). Annealed thermocouples ( $\text{Pt-Pt}_{10}\text{Rh}_{10}$ ) were calibrated frequently with respect to the melting point of pure gold ( $1062.6^\circ\text{C}$ ) and pure synthetic diopside ( $1391.5^\circ\text{C}$ ). Most of the runs near piercing points were performed just after calibration of the thermocouples. The temperatures around these points are estimated to be accurate within  $2^\circ\text{-}3^\circ\text{C}$ . For the other mixtures the accuracy is  $\pm 5^\circ\text{C}$ .

Luth (1967) found that there was loss of potassium in the system  $\text{Mg}_2\text{SiO}_4\text{-KAlSiO}_4\text{-KAlSi}_2\text{O}_6$  when samples were heated in open capsules at high temperatures for a long time. In the present investigation, samples were heated for 3 hours or less at temperatures above  $1350^\circ\text{C}$ . Near the solidus temperatures the maximum duration of a run was 5 hours. Runs of up to one month were made at subsolidus temperatures using both platinum envelope and sealed platinum capsule (2.5 mm outer diameter and 15 mm long). Both ends of the platinum capsule were sealed by natural gas oxygen flame. Sealing process was instantaneous and weighing of the capsule with sample before and after sealing did not show any loss of weight.

Experiments were carried out to determine the percentage loss of potassium

upon heating at various temperatures and for different lengths of time. The results of the mixtures  $LC_{65}Ak_{35}$  and  $LC_{70}Ak_{30}$  are summarized in Table 1.<sup>1</sup> The procedure used was as follows: A platinum envelope was first weighed, then filled with sample, and reweighed. The envelope was crimped at the top, placed in a resistance furnace, and heated as noted in Table 1. The heated sample was then quenched in air and reweighed. Loss of other chemical constituents are also expected in the heating process. Long heating experiments in platinum envelopes show the disappearance of leucite in certain mixtures ( $LC_6Ak_{96}$ ,  $LC_{10}Ak_{90}$ ). This phase is present, when these mixtures are heated for even longer periods in sealed platinum capsules. The loss is therefore interpreted to be primarily due to loss of  $K_2O$ . Table 1 indicates that for samples heated for 3 hours at or below  $1350^\circ C$ , loss is below 1.61 percent of total  $K_2O$ . The loss of  $K_2O$  is estimated to be accurate to  $\pm 1$  percent.<sup>2</sup>

### CRYSTALLINE PHASES

Five crystalline phases were encountered: leucite, forsterite, diopside, akermanite, and monticellite. All phases are solid solutions<sup>1</sup> of varying composition. The indices of refraction and the changes in interplanar spacings of the five crystalline phases are summarized in Table 2.

Leucite<sub>ss</sub> occurs mainly as small, rounded crystals with very low birefringence (.001). Crystals are euhedral with eight sided outline near the liquidus. X-ray study at room temperature indicates that leucite<sub>ss</sub> is tetragonal. Although Bowen and Schairer (1929) reported polysynthetic twinning in this phase, none was seen in the present study.

Forsterite<sub>ss</sub> commonly occurs as stout prismatic crystals with parallel extinction. Near solidus temperatures it forms rounded or anhedral grains. Anhedral forsterite was distinguished by its diffraction peaks at  $32.2^\circ 2\theta$  and  $36.4^\circ 2\theta$  (Cu  $K_\alpha$ ).

Diopside<sub>ss</sub> has stout prismatic form near the liquidus.  $z \wedge c$  in diopside varies between  $38^\circ$ – $42^\circ$ . When coexisting with other phases it commonly forms rounded or anhedral grains, and is then distinguished by its diffraction peak at  $29.78^\circ 2\theta$  (Cu  $K_\alpha$ ).

Akermanite<sub>ss</sub> is prismatic with tetragonal habit near the liquidus; at lower temperature it occurs as rounded grains, and was identified by its low birefringence and diffraction peak at  $31.15^\circ 2\theta$  (Cu  $K_\alpha$ ).

Monticellite<sub>ss</sub> occurs as prismatic crystals with parallel extinction. It was distinguished from forsterite by its lower birefringence and the diffraction peak at  $33.8^\circ 2\theta$  (Cu  $K_\alpha$ ).

<sup>1</sup> In the following passages subscript 'ss' is used to designate solid solution.

<sup>2</sup> To obtain a copy of the experimental data summarized in Tables 1 through 7, order NAPS Document Number 01804. The present address is National Auxiliary Publications Service of the A.S.I.S., c/o CCM Information Corporation, 866 Third Avenue, New York, New York 10022, and price \$2.00 for microfiche or \$5.00 for photocopies, payable in advance to CCMIC/NAPS. Check a recent issue of the journal for current address and price.

In addition to these five crystalline phases, wollastonite<sub>ss</sub> was found in three mixtures (Lc<sub>3</sub>Ak<sub>97</sub>, Lc<sub>5</sub>Ak<sub>95</sub>, and Lc<sub>10</sub>Ak<sub>90</sub>) at and below 1280° ± 10°C. Crystals of wollastonite<sub>ss</sub> and diopside<sub>ss</sub> were observed within large crystals of akermanite<sub>ss</sub> in these mixtures at and below 1240°C.

#### PHASE DIAGRAMS AND QUENCHING DATA

The following "binary" and "ternary" joins were studied in the present investigation; leucite–akermanite, diopside–akermanite–leucite, forsterite–diopside–leucite, and forsterite–akermanite–leucite. A join of composition diopside–akermanite–leucite with 3 percent forsterite was also investigated to determine the temperature and approximate composition of the five-phase piercing point.

##### *Diopside–Akermanite–Leucite*

The join diopside–akermanite was studied by Ferguson and Merwin (1919) and later by Kushiro and Schairer (1964) who suggested that the join has a eutectic at Di<sub>59</sub>Ak<sub>41</sub> and 1361 ± 1°C. This join is not binary because Schairer, Yoder and Tilley (1967) found that pure akermanite is stable only above 1385°C. Below 1345° ± 10°C pseudo-wollastonite and wollastonite<sub>ss</sub> are present with akermanite. Akermanite coexists with diopside and wollastonite<sub>ss</sub> below 1240° ± 10°C. The eutectic of Kushiro and Schairer (1964) is therefore a piercing point. These investigators established that diopside incorporates about 5 weight percent of akermanite as solid solution at 1355°C. Schairer, Yoder, and Tilley (1967) made a mixture of composition Ak<sub>95</sub>Di<sub>5</sub> and studied the solubility of diopside in akermanite. They found that at atmospheric pressure and 1350°C akermanite incorporates less than 5 percent and probably very little diopside.

The join diopside–leucite was worked out by Bowen and Schairer (1929), who showed it to be binary with a eutectic at Di<sub>61.5</sub>Lc<sub>38.5</sub> and 1302° ± 2°C. Two mixtures (Di<sub>95</sub>Lc<sub>5</sub> and Di<sub>97</sub>Lc<sub>3</sub>) were prepared along the join diopside–leucite to determine the amount of leucite incorporated in diopside. The mixtures were melted to glasses and crystallized at 1000°C. X-ray and/or microscopic study of the starting materials of these two mixtures show the presence of diopside and leucite. Crystallized samples were heated at 1285°C in different capsules for 3 hours and 19 hours. Microscopic observation shows well-distributed inclusions of tiny rounded tetragonal leucite crystals (nearly isotropic) within diopside, suggesting that diopside does not incorporate even 3 weight percent of leucite at 1285°C.

Twelve mixtures were prepared to study the join leucite–akermanite.

Experimental results are summarized in Table 3. The equilibrium diagram is shown in Figure 2. Near the akermanite-rich side of the system akermanite<sub>ss</sub> and leucite<sub>ss</sub> coexist with diopside<sub>ss</sub> and wollastonite<sub>ss</sub>. The join is thus not binary. The piercing point occurs at Lc<sub>50.5</sub>Ak<sub>49.5</sub> and 1297° ± 3°C.

X-ray diffraction patterns of leucite from all the mixtures, crystallized at 1275°C for 3 hours show that the reflection (004) is slightly shifted (Table 2) from that of pure leucite, suggesting that it is a solid solution. Leucite is known to form a complete series of solid solution with the compound K<sub>2</sub>O·MgO·5SiO<sub>2</sub> (Roeder, 1951; Schairer, 1948).

Pure akermanite is stable only above 1385°C (Schairer, Yoder and Tilley, 1967); this phase therefore is a solid solution. The temperature at which akermanite<sub>ss</sub> is joined by leucite<sub>ss</sub> increases from Lc<sub>40</sub>Ak<sub>60</sub> to Lc<sub>20</sub>Ak<sub>80</sub>. A mixture of composition Lc<sub>3</sub>Ak<sub>97</sub> heated at 1280°C for 2 hours shows well distributed inclusions of leucite<sub>ss</sub> and wollastonite solid solution with akermanite<sub>ss</sub>, which suggests that akermanite does not incorporate even 3 weight percent of leucite as solid solution. Exact composition of akermanite<sub>ss</sub> was not established.

Five mixtures of compositions Lc<sub>5</sub>Ak<sub>95</sub>, Lc<sub>10</sub>Ak<sub>90</sub>, Lc<sub>20</sub>Ak<sub>80</sub>, Lc<sub>40</sub>Ak<sub>60</sub>, and Lc<sub>70</sub>Ak<sub>30</sub> were chosen to study the join in the subsolidus region. In their study of the thermal behavior of pure synthetic akermanite, Schairer, Yoder, and Tilley (1967) found well distributed inclusions of diopside and wollastonite<sub>ss</sub> within melilite at and below 1240 ± 10°C. The above-mentioned mixtures were therefore heated at 1240°C in

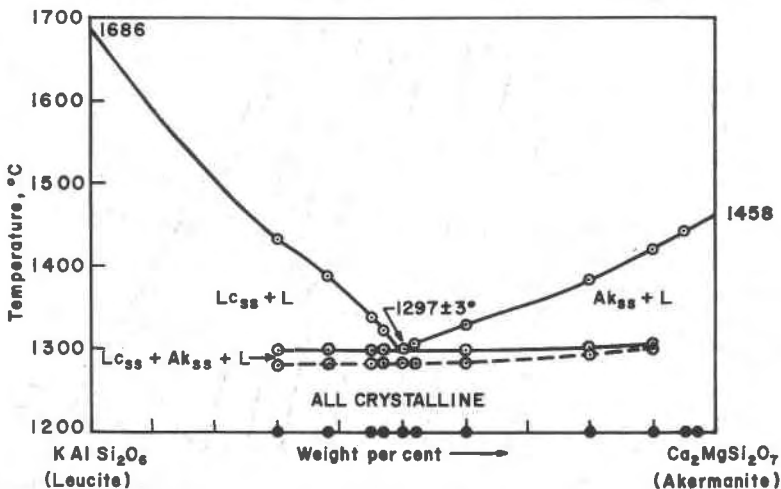


Fig. 2. Phase equilibrium diagram of the system leucite-akermanite.

sealed platinum capsules for 24 days and in platinum envelopes for 28 days to check the possibility of the presence of diopside and wollastonite<sub>ss</sub> in the subsolidus region of the system leucite-akermanite.

X-ray and microscopic observation of the samples heated at 1240°C for 24 days in sealed capsules show that leucite<sub>ss</sub>, diopside<sub>ss</sub>, and wollastonite<sub>ss</sub> coexist with akermanite<sub>ss</sub> in the mixtures Lc<sub>5</sub>Ak<sub>95</sub> and Lc<sub>10</sub>Ak<sub>90</sub>, whereas the other mixtures contained only leucite<sub>ss</sub> and akermanite<sub>ss</sub>.

X-ray and microscopic study of the samples heated in platinum envelopes for 28 days show that mixtures of compositions Lc<sub>20</sub>Ak<sub>80</sub>, Lc<sub>40</sub>Ak<sub>60</sub>, and Lc<sub>70</sub>Ak<sub>30</sub> contain diopside<sub>ss</sub>, leucite<sub>ss</sub>, and akermanite<sub>ss</sub>, whereas the mixtures Lc<sub>10</sub>Ak<sub>90</sub> and Lc<sub>5</sub>Ak<sub>95</sub> contain akermanite<sub>ss</sub> and diopside<sub>ss</sub>. The samples heated for 28 days were crystallized at 1280°C in platinum envelopes for 21 hours. All the samples contained diopside<sub>ss</sub> and akermanite<sub>ss</sub> with or without leucite<sub>ss</sub> as indicated earlier. Absence of leucite<sub>ss</sub> in the mixtures Lc<sub>10</sub>Ak<sub>90</sub> and Lc<sub>5</sub>Ak<sub>95</sub> is attributed to the loss of potassium. Experimental results of the mixtures heated for 28 days in platinum envelopes may not be reliable, because the loss of

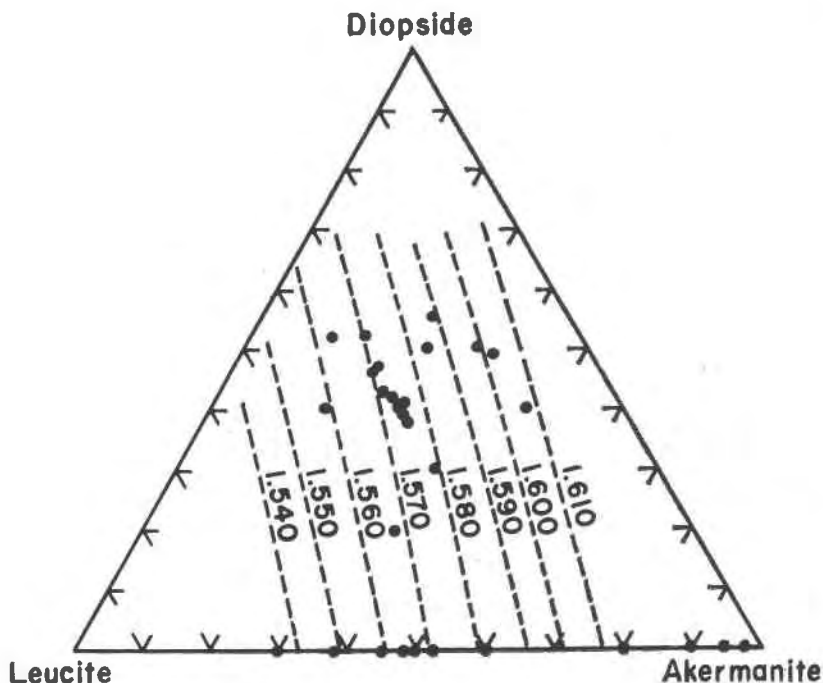


FIG. 3. Isofract lines in the system diopside-akermanite-leucite.

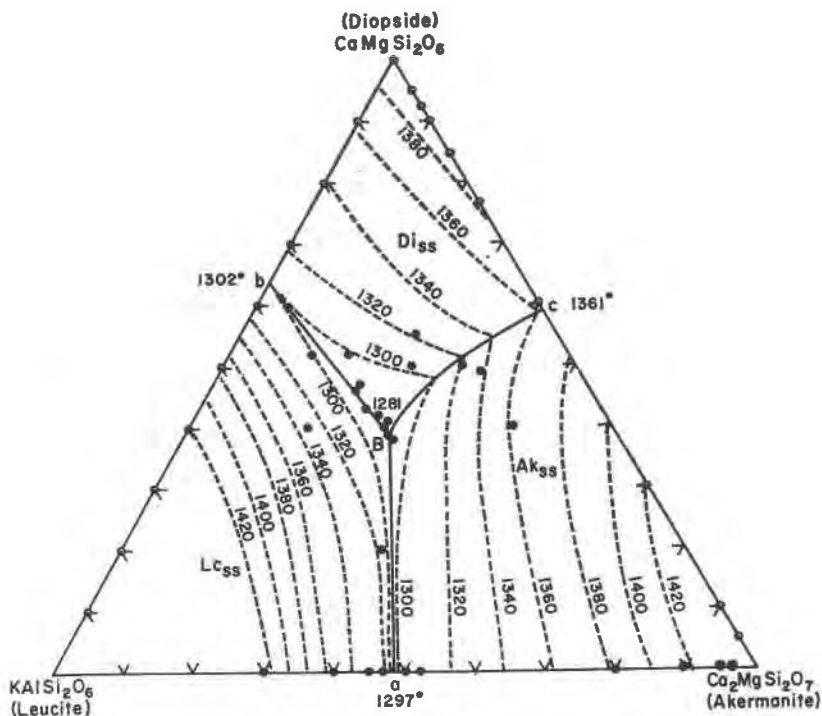


FIG. 4. Phase equilibrium diagram of the system diopside-akermanite-leucite.

potassium would produce excess alumina and silica which may react with akermanite to produce diopside<sub>ss</sub>.

Nineteen mixtures were made within the join diopside-akermanite-leucite. Isofract lines are presented in Figure 3. Figure 4 shows the equilibrium diagram. Experimental results are summarized in Table 4.<sup>1</sup> The solid lines divide the primary phase fields for different phases. Solid dots represent the mixtures made in the present study. The dotted lines represent the isotherm. At point B ( $\text{Di}_{39}\text{Ak}_{29}\text{Lc}_{32}$ ) diopside<sub>ss</sub> + akermanite<sub>ss</sub> + leucite<sub>ss</sub> + liquid coexist in equilibrium at  $1281^\circ \pm 3^\circ\text{C}$ . Wollastonite<sub>ss</sub> is absent on the liquidus of this join. As the compositions of the phases do not lie within the join, the system is not ternary. Point B is therefore a piercing point and the lines dividing the primary phase fields are traces of divariant surfaces. In addition to akermanite, diopside may contain some alumina as solid solution. Clark, Schairer, and de Neufville (1962, p. 62) studied the changes in

<sup>1</sup> In the tables from 3 to 7, B.M. refers to beginning of melting.



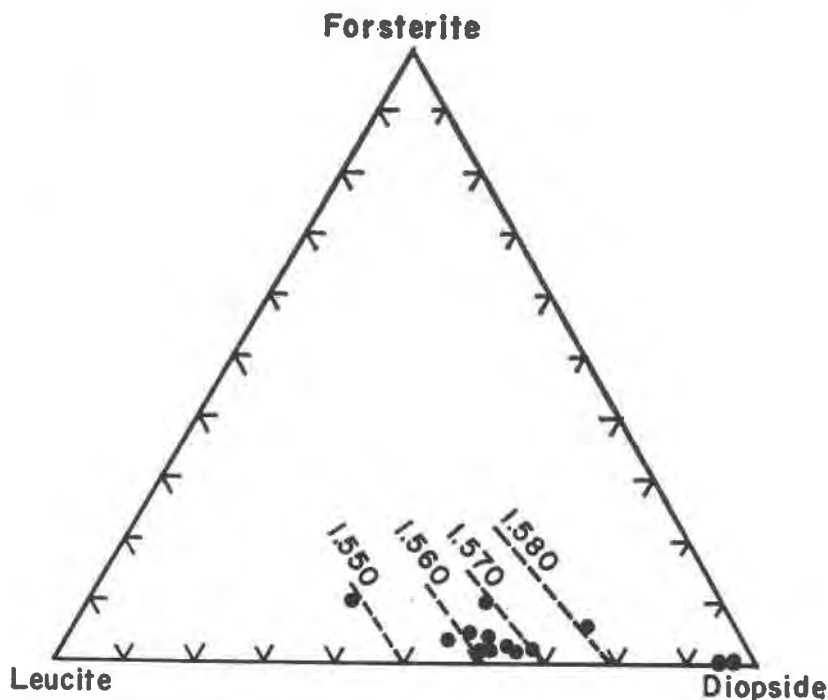


FIG. 5. Isofrac lines in the system forsterite-diopside-leucite.

the interplanar spacings of diopside solid solution containing variable amount of  $\text{CaAl}_2\text{SiO}_6$  molecule. They used nine reflections which were sharp, strong, and sensitive to solid solution. Of these reflections only (221) of diopside<sub>ss</sub> ( $\text{Di}_{40}\text{Ak}_{28}\text{Lc}_{32}$ ) was found in the join under consideration to be deviated by  $0.008\text{\AA}$  (table 2) when compared with that of pure diopside (Table 2). Eight other reflections do not show any noticeable changes. The measurement is estimated to be accurate to  $\pm 0.005\text{\AA}$ . De Neufville and Schairer (1962, p. 58) found that changes in the reflection (211) of melilite can be used to determine the changes in the composition of this phase in the system diopside- $\text{CaAl}_2\text{SiO}_6$ . In the join diopside-akermanite-leucite, reflection (211) of melilite ( $\text{Di}_{41}\text{Ak}_{28}\text{Lc}_{31}$ ) is shifted by  $0.007\text{\AA}$ , when compared to that of pure synthetic akermanite. Study of the changes in the interplanar spacings of leucite<sub>ss</sub> containing variable amount of  $\text{K}_2\text{O}\cdot\text{MgO}\cdot 5\text{SiO}_2$  molecule has not been made. Reflection (004) of leucite<sub>ss</sub> was found to be shifted by  $0.022\text{\AA}$  ( $\text{Di}_{38}\text{Ak}_{30}\text{Lc}_{32}$ ; table 2), when compared to that of pure leucite, which suggests that this phase is a solid solution. Table 4 shows that a liquid phase persists  $40^\circ\text{C}$  below  $1281^\circ \pm 3^\circ\text{C}$  (point B).

Persistence of a liquid phase may be explained by: (a) short duration of experimental runs to minimize loss of potassium, (b) presence of solid solution in all three phases, and (c) viscosity of the melt.

### *Forsterite-Diopside-Leucite*

Of the three bounding "binary" joins, diopside-leucite was previously discussed. The join forsterite-diopside was studied by Bowen (1914) and later by Kushiro and Schairer (1963). The latter workers found it to be pseudobinary because forsterite contains monticellite in solid solutions. They established that diopside can contain about 5 weight percent of forsterite in solid solution at least at temperatures above 1300°C. A piercing point occurs at  $1389 \pm 1^\circ$  and a composition of  $\text{Fo}_{11}\text{Di}_{89}$ . Schairer (1954) prepared two mixtures along the join forsterite-leucite and suggested that the system is binary with the eutectic point at  $1493^\circ \pm 5^\circ\text{C}$  and at a composition of  $\text{Fo}_{21}\text{Lc}_{79}$ .

Eleven mixtures were made within the "ternary" join. Isofract lines are presented in Figure 5. As the number of mixtures made for the study of this join is small, isofract lines presented in Figure 5 may not be a satisfactory test of the homogeneity of the glasses. However agreement of data with those of nearby mixtures and good distribution of the phases in the glasses suggest that glasses were homogeneous. Figure 6 shows the equilibrium diagram for the join forsterite-diopside-leucite. The results are summarized in Table 5. At point C ( $\text{Fo}_3\text{Di}_{60}\text{Lc}_{37}$ ) forsterite<sub>ss</sub> + diopside<sub>ss</sub> + leucite<sub>ss</sub> + liquid coexist in equilibrium at  $1296^\circ \pm 3^\circ\text{C}$ . Diopside may contain some alumina as solid solution. As the compositions of all the phases do not lie in this join, it is not ternary. Indices of refraction and the interplanar spacings of the phases are given in Table 2. Table 5 shows that the lowest solidus temperature is  $1245^\circ \pm 10^\circ\text{C}$ .

### *Forsterite-Akermanite-Leucite*

Akermanite-leucite and forsterite-leucite have already been discussed. Information on forsterite-akermanite is available from the system  $\text{CaO-MgO-SiO}_2$  (Ferguson and Merwin, 1919). This join cuts 1430°C. Schairer, Yoder, and Tilley (1967) made two mixtures the primary phase volume of monticellite<sub>ss</sub> and is pseudobinary above ( $\text{Fo}_{98}\text{Ak}_2$  and  $\text{Fo}_{95}\text{Ak}_5$ ) in the join forsterite-akermanite and established that akermanite contains less than 2 percent forsterite as solid solution at 1340°C. Ferguson and Merwin (1919) suggested that forsterite and monticellite are solid solutions.

Nineteen mixtures were made in the join forsterite-akermanite-leucite. Experimental data for the system are summarized in Table 6.

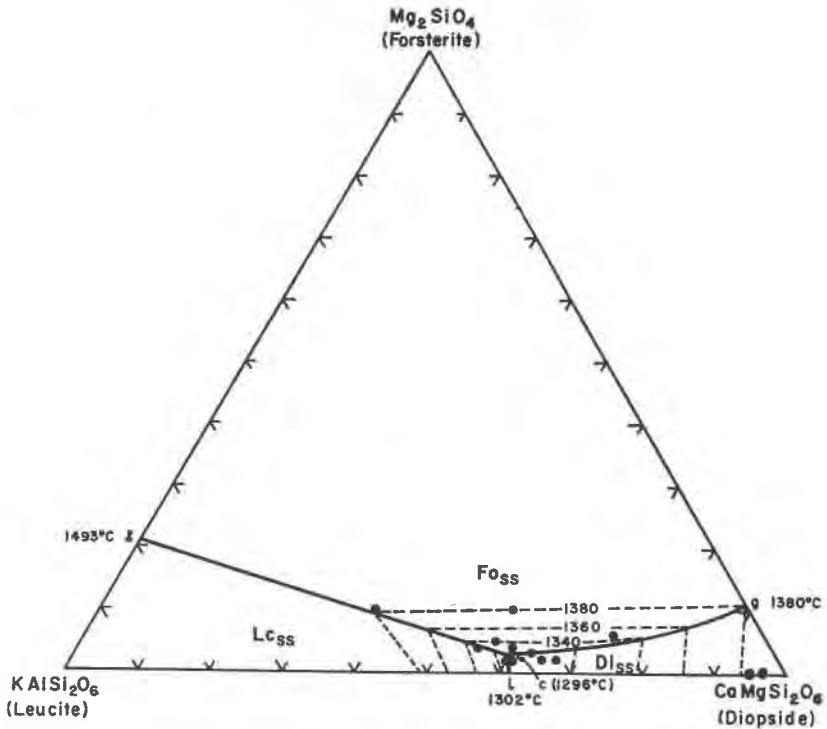


FIG. 6. Phase equilibrium diagram of the system forsterite-diopside-leucite.

Isofract lines are presented in Figure 7. Figure 8 shows that the join is pseudoternary at least above  $1428^{\circ}\text{C}$  and cuts through the primary phase volume of monticellite<sub>ss</sub>. The join has two piercing points at  $\text{Fo}_{17}\text{Ak}_{78}\text{Lc}_5$  (H) and  $1428^{\circ} \pm 3^{\circ}\text{C}$  where forsterite<sub>ss</sub> + monticellite<sub>ss</sub> + akermanite<sub>ss</sub> + liquid coexist, and at  $\text{Fo}_9\text{Ak}_{43.5}\text{Lc}_{47.5}$  and  $1286^{\circ} \pm 3^{\circ}\text{C}$  (G) of which the assemblage is forsterite<sub>ss</sub> + akermanite<sub>ss</sub> + leucite<sub>ss</sub> + liquid. Monticellite<sub>ss</sub> reacts with liquid and is completely eliminated at  $1410^{\circ} \pm 5^{\circ}\text{C}$ ; it does not coexist with leucite<sub>ss</sub>. Disappearance of monticellite can be ascribed to its reaction with liquid to produce akermanite. Forsterite contains monticellite in solid solution. X-ray diffraction patterns of forsterite<sub>ss</sub> from a mixture ( $\text{Fo}_{17}\text{Ak}_{81}\text{Lc}_2$ ) crystallized at  $1400^{\circ}\text{C}$ , show that reflection (222) is at  $1.762 \pm 0.005 \text{ \AA}$  (table 2). This value suggests, according to Rieker and Osborn's (1954) diagram that forsterite contains considerable monticellite as solid solution. Reflection (222) of monticellite from the same mixture is at  $1.81 \text{ \AA}$  whereas the same reflection for pure synthetic monticellite is at

$1.819 \pm 0.005 \text{ \AA}$  which may indicate that monticellite contains forsterite in solid solution.

*Forsterite-Diopsid-Akermanite*

This join was studied by Ferguson and Merwin (1919). They found that at  $\text{Fo}_{0.17}\text{Ak}_{77}\text{Di}_6$  (estimated from their system  $\text{CaO-MgO-SiO}$ ) and  $1430^\circ\text{C}$ , forsterite + monticellite + akermanite + liquid are in equilibrium. The eutectic point occurs at  $\text{Fo}_{8.5}\text{Di}_{50}\text{Ak}_{41.5}$  where forsterite + diopside + akermanite + liquid coexist at  $1357^\circ\text{C}$ . In this join monticellite has a reaction relation with the liquid, and the final assemblage is forsterite + diopside + akermanite.

*The join diopside-akermanite-leucite with 3 percent forsterite*

The join was studied to determine the temperature of the five-phase (forsterite<sub>3</sub> + diopside<sub>3</sub> + akermanite<sub>3</sub> + leucite<sub>3</sub> + liquid) piercing point in the system forsterite-diopside-akermanite-leucite. Considerations of the bounding joins of the tetrahedron suggest that the piercing point lies very close to the basal plane diopside-akermanite-

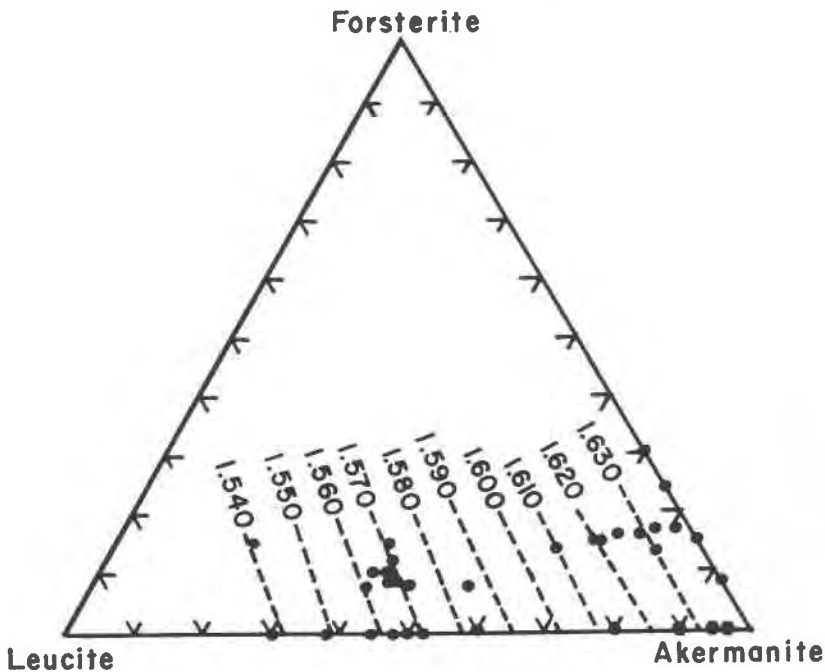


FIG. 7. Isofract lines in the system forsterite-akermanite-leucite.

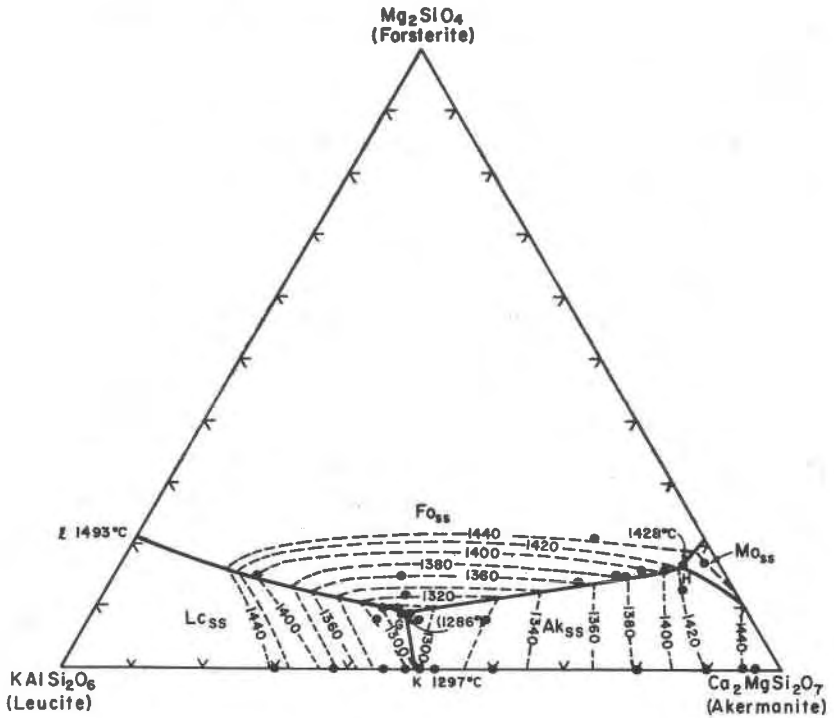


FIG. 8. Phase equilibrium diagram of the system forsterite-akermanite-leucite.

leucite and probably contains less than 5 percent forsterite. Accordingly, the join diopside-akermanite-leucite with 3 percent forsterite was studied. Twelve glasses were made and crystallized. The quenching results are summarized in Table 7. The equilibrium diagram for the join is represented by Figure 9. At K ( $\text{Fo}_3\text{Di}_{55}\text{Ak}_9\text{Lc}_{33}$ ) forsterite<sub>ss</sub> + diopside<sub>ss</sub> + leucite<sub>ss</sub> + liquid are in equilibrium at  $1295^\circ \pm 3^\circ\text{C}$ , and at L ( $\text{Fo}_3\text{Di}_{41}\text{Ak}_{24}\text{Lc}_{32}$ ) diopside<sub>ss</sub> + akermanite<sub>ss</sub> + leucite<sub>ss</sub> + liquid coexist in equilibrium at  $1274^\circ\text{C}$ , below which there is a liquid phase which does not freeze up until  $1255^\circ\text{C}$ . As the compositions of the crystalline phases lie outside the join, and there is a liquid phase below  $1274^\circ\text{C}$ , the system forsterite-diopside-akermanite-leucite is not quaternary. Optical and X-ray study suggests that the phases in this join contain only a small amount of solid solution (Table 2). The behavior of the join approximates that of a quaternary system and points K and L behave as piercing points. From these two points liquid continues to crystallize along two different paths and at  $1274^\circ\text{C}$  the five-phase assemblage of forsterite<sub>ss</sub>, diopside<sub>ss</sub>, akermanite<sub>ss</sub>, leucite<sub>ss</sub>,

and liquid is reached. Below 1274°C crystallization continues as the liquid moves towards the lowest melting point. Study of this join suggests that the lowest melting point of the tetrahedron, forsterite–diopside–akermanite–leucite lies close to the basal plane diopside–akermanite–leucite. The final composition of the liquid thus lies near this plane.

#### PETROLOGICAL SIGNIFICANCE OF THE SYSTEM FORSTERITE–DIOPSIDE–AKERMANITE–LEUCITE

The system forsterite–diopside–akermanite–leucite is a simplified model in which compositions of potassium-bearing mafic and ultra-mafic rocks can be plotted (Figure 1). Study of the system helps to understand the relation between these rocks.

#### Melilite leucitite

Figure 4 shows that melilite leucitite (piercing point B) can be derived from either melilite itelite (aB), leucitite (bB) or melilitite (cB). Natural melilitites contain a considerable amount of sodium as soda melilite in solid solution (Schairer and Yoder, 1964), however they are poor in  $K_2O$ . The melilitites referred to here have compositions lying in the tetrahedron forsterite–diopside–akermanite–leucite. At

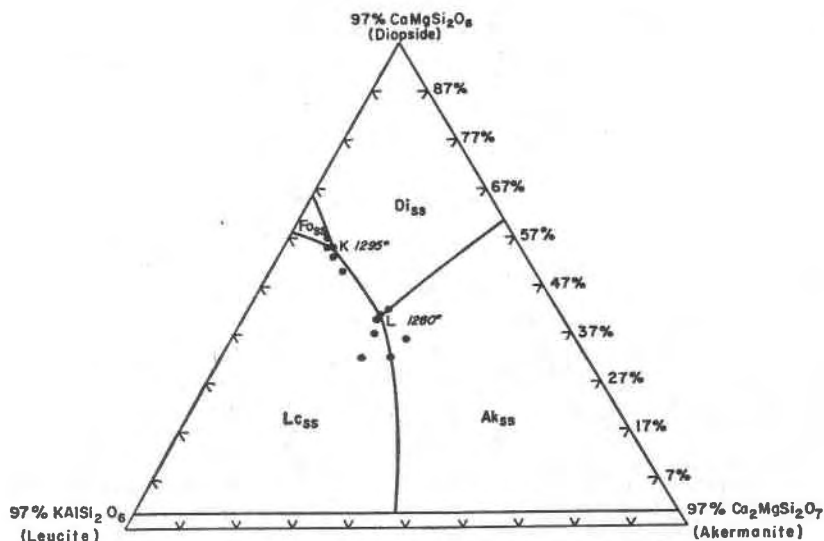
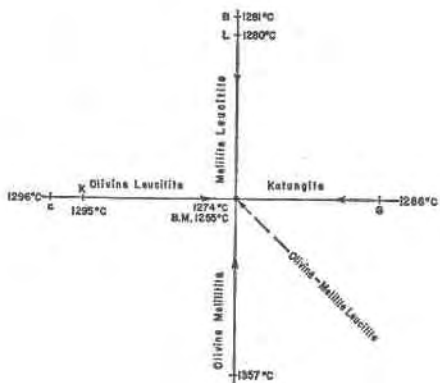
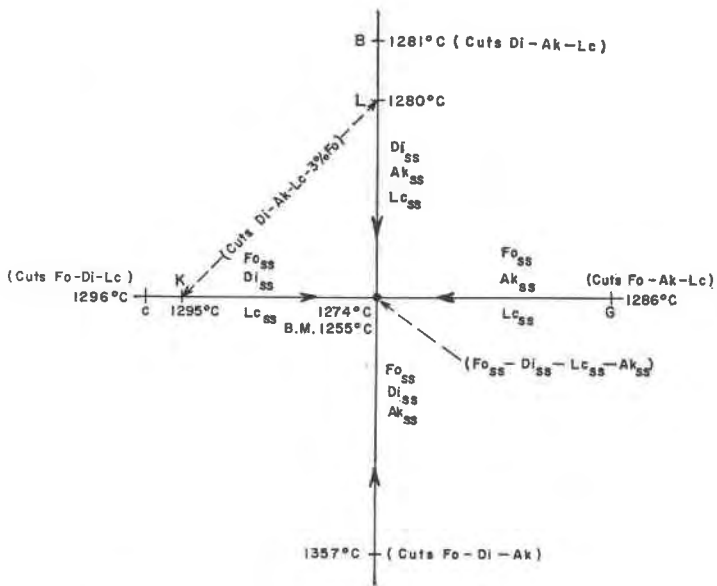


FIG. 9. Phase equilibrium diagram of the system diopside–akermanite–leucite with three per cent forsterite.



(a)



(b)

FIG. 10. (a) Flowsheet diagram of the system forsterite-diopside-akermanite-leucite. (b) Rock nomenclature diagram of the same system.

Capo di Bove in Italy, Washington (1906) noted the sporadic occurrence of leucitite grading into melilite leucitite (cecilite). Results of the system diopside-akermanite-leucite indicate that melilite leucitite may be derived from leucitite magma.

#### *Olivine leucitite*

Figure 6 shows that olivine leucitite (piercing point C) can be derived from either potassium-rich olivine pyroxenite (gc), olivine itaite (lc), or leucitite (ic). Olivine pyroxenites ordinarily contain very low amounts of  $K_2O$ . The olivine pyroxenites referred to here have compositions lying in the tetrahedron forsterite-diopside-akermanite-leucite. In the Bufumbira region of Uganda, Holmes (1937) described a lava flow of olivine leucitite (ugandite). At Katunga in Uganda, he noted the close association of rocks such as leucitite and olivine leucitite and also reported the complete gradation of these rocks to pyroxene-rich leucitite, which in turn grades into pyroxenite. This field evidence and the results on the system forsterite-diopside-leucite suggest that an olivine-leucitite magma can be a derivative of a potassium-rich olivine pyroxenite liquid. Generation of such a liquid is possible by the melting of mica-bearing ultrabasic rocks (Yagi and Matsumoto, 1966).

#### *Katungite*

Phase equilibria study of the system forsterite-akermanite-leucite suggests that a katungite magma (piercing point G, Figure 8) can be derived from melilite itaite (kG), olivine itaite (lG), or alnoite (HG). Field evidence of a massive flow of katungite lava was reported by Holmes (1937) from Ankole in Uganda, where it occurs in close association with alnoite. On the basis of the present experimental study it appears possible that katungite may have been derived from alnoite, which itself may have originated from monticellite alnoite.

#### *Olivine melilitite*

Reference to Ferguson and Merwin's (1919) diagram forsterite-diopside-akermanite shows that an olivine melilitite may be produced from a peridotite, melilitite, or an alnoite magma.

### CONCLUSIONS

The system forsterite-diopside-akermanite-leucite is a pseudoquaternary system that is a part of the quinary system  $K_2O-Al_2O_3-CaO-MgO-SiO_2$ . The compositions of all the crystalline phases lie outside the tetrahedron. The amount of solid solution in all these phases is small. The system can thus be treated as a quaternary. The com-



plete course of crystallization of liquid is summarized by a flowsheet diagram, as shown in Figure 10a. Figure 10b, a rock nomenclature diagram corresponding to the flowsheet, shows that olivine-melilite leucitite can be produced from either melilite leucitite, olivine leucitite, katungite, or olivine melilitite. Schairer and Yoder (1964) studied the system nepheline-forsterite-larnite-silica, and showed that parental liquids for the sodium-rich undersaturated part may be represented by either olivine melilitite or olivine nephelinite. Olivine-melilite nephelinite is produced from liquids of these compositions. It appears that rocks called olivine melilitite can differentiate toward either olivine-melilite nephelinite or olivine melilite leucitite. The  $\text{Na}_2\text{O}/\text{K}_2\text{O}$  ratio of the olivine melilitite parent probably controls the subsequent course of crystallization. If the liquid is potassium-rich, leucite-bearing mafic and ultramafic rocks would be produced. If the liquid is sodium-rich, nepheline-bearing mafic and ultramafic rocks would result. Further studies on the "effect" of sodium and potassium content of under-saturated mafic and ultramafic rocks are in progress.

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