Compositional variation in late-stage differentiates in mare lavas, as indicated by silicate melt inclusions

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Abstract—Approximately 1000 electron microprobe analyses (484 previously unpublished) of silicate melt inclusions and of their host and associated phases in lunar mare basalts are presented (in archival form). Only about 100 analyses are of inclusions in olivine—the rest are mainly from late stages of crystallization, and hence provide some insight into the late differentiation trends for these magmas. There are three compositional types of late-stage melts: immiscible “high-Fe” and “high-K” melts, and a possibly contemporaneous but enigmatic “low-K” melt. The factors affecting the interpretation of these data are reviewed, particularly the fragmental nature of the record and crystallization of the trapped melt. The compositional data differ from sample to sample, thus permitting a grouping and recognition of some rock types; they verify but do not explain the previously reported strongly bimodal distribution of $K_2O$ values for inclusions in ilmenite; and they delineate some differentiation trends (e.g., increasing $SiO_2$, $K_2O$, and $FeO/FeO + MgO$), but some unanswered questions remain. Thus, silicate liquid immiscibility seems to occur at ~2% $K_2O$, but the stage cannot be specified exactly, and the origin of the low-K, high-Si melt inclusions in ilmenite reported earlier remains obscure.

INTRODUCTION

We have made 977 electron microprobe analyses of melt inclusions and of their host and associated phases in lunar mare basalts, of which 484 are new; the rest have been published (Roedder and Weiblen, 1970, 1971, 1972a,b, 1974, 1975; Powell and Weiblen, 1972; Weiblen and Roedder, 1973). As a result of the sample allotments, this work covered a wide variety of mare samples; the main goal was to investigate the liquid line of descent for lunar mare magmas through analyses of silicate melt inclusions.

Although volumetrically insignificant on the moon, the late-stage differentiates are extremely important in problems of lunar chronology. The “magic” component in lunar soils has been discussed by several authors (e.g., Papastassiu et al., 1970; Evensen et al., 1974). Evensen (pers. comm., 1977) has reviewed the data on this “magic” component and shows that: (1) it is found in all soils sampled on the moon, (2) it consists of varying proportions of two fractions, one derived from the local rocks, and the other derived from rocks older than the mare lavas, (3) it was transported and mixed in local soils as fragmental material, and (4) it occurs in increasing amounts in the smaller size fractions of the soils. Thus beyond the considerations of petrologic curiosity, it is important to characterize and understand the origin of the mesostasis in the
mare lavas. For the latter problem, we must know the details of the liquid line of
descent in the mare lavas which leads to these compositions. The recognition of
possible early silicate immiscibility (>20% melt) by Hollister and Crawford
(1977) in an Apollo 15 sample gives additional impetus to define the liquid line of
descent in mare lavas.

In this paper, we review the factors that must be considered in interpreting
data from silicate melt inclusions, present and discuss the new analytical data,
and attempt a synthesis of the old and new data. In particular, we characterize
the late-stage mesostasis and discuss problems of its origin. In a companion
paper (Weiblen, 1977), the early part of the liquid line of descent is examined in light
of data from melt inclusions in olivine.

**INTERPRETATION OF DATA FROM SILICATE MELT INCLUSIONS**

Although silicate melt inclusions provide some types of data not obtainable
from any other source, they are not a panacea for problems in petrogenesis. A
series of factors that can cause difficulty or ambiguity in interpretation has been
discussed by Roedder (1976) and, for the most part, need not be repeated here.
However, three are particularly pertinent to the lunar data:

1. *The fragmental nature of the record*

   In addition to the problem inherent in the nature of the lunar rock sampling,
it is necessary to remember that inclusions represent defects in the otherwise
orderly process of growth of host crystals. As such, they can form by many
different mechanisms, but in the lunar samples, most melt inclusions have been
trapped by three processes: *First*, early skeletal or tubular growth may be
enclosed by later solid growth, trapping melt. This may result in a single,
relatively large inclusion, as in some “cored phenocrysts” of plagioclase and
pyroxene, where the composition, nucleation, and growth rate were favorable
(e.g., see Donaldson *et al.*, 1975), but such large inclusions are relatively
uncommon. Lunar olivines, however, commonly contain one or more inclusions
that have an almost random spatial distribution, and have no noticeable bias
toward occurrence in the cores. We presume that these inclusions are also the
result of skeletal growth on crystal edges at various stages, as is sometimes
noted in terrestrial olivine phenocrysts. Some terrestrial olivines commonly have
axially arrayed inclusions (Roedder and Weiblen, 1971, their Fig. 1). We do not
know what variables cause this difference. The only other lunar mineral that has
consistently trapped melt inclusions by skeletal growth is ilmenite, which
sometimes contains as much as 50% extraneous material (e.g., 10047). Most such
inclusions consist of single crystals, optically continuous with nearby crystals
and hence presumably connected with them in the third dimension, but inclusions
representing the trapping of a homogeneous melt are also common in
ilmenite (Roedder and Weiblen, 1970, their Figs. 2 and 4). *Second*, the enclosure
of another crystal by the growing host crystal may trap some melt as well. The
most common examples are melt inclusions attached to spinel euhedra enclosed within early olivine. Third, immiscible liquid droplets present within the liquid from which the host crystal is growing may adhere to the crystal surface and become trapped. By our interpretation, this last process may explain the very common occurrence of a row of tiny inclusions of glass of potash granite composition near the margin of many of the crystals of plagioclase, pyroxene, and other late phases in most of the mare basalts (e.g., Roedder and Weiblen, 1970, their Figs. 14, 15, and 17; 1971, Fig. 21). Earlier crystal growth is free of such inclusions, presumably because the liquid was a single phase at that time. Several features of these inclusions seem to preclude an origin for them based on a sudden change in conditions causing skeletal growth and hence a row of inclusions (e.g., there is no preferential occurrence of such rows on the relatively fastest growing faces), and the chemical compositions of some of the host phases, such as ilmenite, which could hardly grow rapidly from a melt of potash granite composition.

We have previously shown (Weiblen and Roedder, 1976) that the crystallization sequence in mare basalts is spinel, olivine, ilmenite, and then plagioclase plus pyroxene, leading to a late-stage separation of two immiscible melts. We have found a few melt inclusions in spinel, and many in olivine, giving a reasonable sampling of melts through the early stages of crystallization. However, the bulk of the crystals formed from a mare magma are pyroxene and plagioclase, and except for the last stages of growth of these two, when two liquids were present, they seldom have trapped analyzable inclusions. Ilmenite crystallizes over a wide range, and contains abundant inclusions, but most of these are in the part that crystallized late, when the residual melt contained 60% or more of SiO₂. Therefore, melt inclusions present us with an incomplete record of the sequence of melts at best, since most are incorporated in either very early or very late phases.

A dilemma concerning the origin of the two grossly different inclusion compositions found in ilmenite (Roedder and Weiblen, 1975) is still unresolved. Both contain approximately 75% SiO₂, but whereas one group contains ~6% K₂O, the other averages near to 0% K₂O.

2. The possible effects of cooling rate on late-stage melt composition

J. E. Quick (pers. comm., 1977) has suggested that the composition of late-stage, high-silica melts might be a function, in part, of the cooling rate. Grove and Bence (1977) showed by experimental runs on a synthetic analog of the Apollo 15 quartz-normative basalt 15597 that the sequence of phases, and their compositions (particularly pyroxene) changed significantly with cooling rate, from 0.5 to 150°C/hr. These differences affected the distribution of both major and minor elements between crystals and liquid, and hence could be expected to affect the composition of the late-stage liquids.

The optimum test of this effect on late-stage melts would be to compare the late-stage melt compositions in pairs of rocks of identical bulk composition but
differing only in cooling rate. There is a range of grain sizes among the samples we have examined, presumably covering a rather broad range of cooling rates (Usselman et al., 1975), but we cannot know whether the cooling rate has been a significant factor in the late-stage melt composition since the bulk compositions are far from identical. A high cooling rate would also favor an increased compositional range for the residual melts due to local crystal-liquid equilibria.

3. **Crystallization of the trapped melt**

Except for those with very high-silica content, most lunar inclusions have crystallized to some degree. If this crystallization involves only deposition of the host mineral on the walls, or epitaxial growth of a single daughter crystal (e.g., an ilmenite plate on the olivine host (Roedder and Weiblen, 1970), the residual melt represents the original melt minus the amounts of the crystallized phases. The amount of crystallization of the host phase on the walls can be large (~30%; Anderson, 1974). As many crystalline inclusions are essentially opaque, some may have no visible daughter ilmenite or other coarse phases, i.e., none intersecting the polished surface, simply because of the plane of the random cut. As a result of this crystallization, on any type of variation diagram, the composition of the liquid at the time of trapping should fall along the control line between the present composition of the inclusion and the appropriate crystallized phase(s).

If, as is common, the residual melt around the daughter crystal has also crystallized to a feathery intergrowth of pyroxene, etc. (Figs. 1–4), analyses of the mixture will show scatter reflecting this heterogeneity, adding to the scatter from loss due to crystallization of host or daughter phases and from systematic analytical errors (Albee et al., 1977). Sometimes most of the crystals are fine enough (≤2 μm; Figs. 1 and 3) for a defocused beam (10–20 μm) to give a reasonably good average analysis, but if the crystals are coarse (Figs. 2 and 4), and/or the inclusion is small, no analytical method on a single random section can yield a valid analysis. Unfortunately, in most inclusions analyzed, at least some of the crystals exceeded 2 μm.

**New data obtained**

To fill gaps in what must remain at best a fragmental record because of the nature of the original sampling on the moon, and to minimize the difficulties of interpretation inherent in the above, we selected additional slides that were optimum for the purpose, and made 484 new analyses. Most of these were of inclusions in olivine and ilmenite, in Apollo 11 low-K basalts 10020,51,−82, and −107; 10044,41; 10047,11; and 10062,39; Apollo 12 low-Ti olivine basalt 12035,23 and unclassified basalt 12012,7; Apollo 15 low-Ti olivine basalt 15545,64 and unclassified basalt 15598,11; and unclassified Apollo 17 basalts 71135,28 and 78505,65 (classifications from Papike et al., 1976). Some of the results from these new analyses were presented in graphical form earlier (Weiblen and Roedder,
Compositional variation in late-stage differentiates in mare lavas

Figs. 1–4. Fine- and coarse-grained crystalline silicate melt inclusions in olivine and ilmenite from the samples indicated, in reflected plain light, all at the same magnification. (Scale bar = 20 μm.)

Fig. 1. Typical crystallized inclusion in olivine of low-Ti olivine basalt 12035. An epitaxial daughter crystal of ilmenite (vertical bar) is embedded in a granular matrix consisting of feathery pyroxene crystals in glass(?). Analysis of this matrix (Table 1, tape no. 77) shows 53.9% SiO₂ and 0.15% K₂O.

Fig. 2. Coarsely crystallized inclusion in olivine. This might well also have a hidden epitaxial daughter crystal like that in Fig. 1. The coarseness of the crystals would make defocused-beam analysis rather inaccurate without corrections (Albee et al., 1977), even if this particular section were truly representative.

Figs. 3–4. Melt inclusions in ilmenite from low-K basalt slide 10047,11. The inclusion in Fig. 3 apparently consists of essentially all glass, and a few feathery crystals, whereas that in Fig. 4 apparently nucleated pyroxene earlier in the cooling cycle and hence is now rather coarse.

1976), but the full analyses are given for the first time as Table 1¹. The methods of analysis and data reduction are the same as described earlier (Roedder and Weiblen, 1973, p. 1032) and are given in the notes to Table 1. Only about 100 of the analyses in Table 1 are of inclusions in early phases such as olivine; most of the other inclusions analyzed (in ilmenite or interstitial) are high in SiO₂ (>70%) and hence presumably trapped during late-stage crystallization.

¹The 484 new analyses, along with our 1288 previously published analyses of lunar samples, (and including ~770 not directly pertinent to melt inclusions), are listed in Table 1. Each of the 1772 entries has a sequential tape number, a reference number (the unpublished analyses bear reference numbers 261–744), and are grouped into categories based on the nature of the sample analyzed, where published, or both. In order to save space, the table, consisting of 75 pages, has been archived. See NAPS document no. 03095 for 113 pages of supplementary material. Order from ASIS/NAPS c/o Microfiche Publications, P.O. Box 3513, Grand Central Station, New York, NY 10017. Remit in advance for each NAPS accession number. Institutions and organizations may use purchase orders when ordering, however, there is a billing charge for this service. Make checks payable to Microfiche Publications. Photocopies are $28.25. Microfiche are $3.00 each. Outside the United States and Canada, postage is $3.00 for a photocopy or $1.00 for a fiche. The data can also be obtained as a magnetic tape or punched cards from the authors.
OVERALL CHARACTERIZATION OF LATE-STAGE DIFFERENTIATES

It is easy to infer from the references to late-stage “mesostasis” in the literature on the mare basalts that there is essentially only one composition. In the following brief review, we want to emphasize that not only are there three quite distinct major compositional types, which we have termed high-Fe, high-K, and low-K, but that these three types are found in specific textural occurrences, and also reveal compositional variations within each for individual rocks or groups of rocks.

Textural and mineralogical features

The late-stage differentiates occur: (1) as silicate melt inclusions within crystals, (2) as interstitial mesostasis, in the mare basalts, and (3) as clasts in breccias. All three of these types of materials may contribute to the “exotic” component in lunar soils, and it is the late-stage differentiates that have yielded the minor but geochemically important lunar phases such as zirconolite, pyroxferroite, tranquillityite, apatite, whitlockite, and celsian, as well as the less exotic cristobalite, fayalite, and iron-rich pyroxenes. As a result of the absence of volatiles and hence high viscosity of these melts (other than the high-Fe), and the relatively high cooling rates, an appreciable fraction of these late-stage differentiates has remained as glass. In view of the possible significance, the partitioning of geochemically important elements between these various phases, and the melt, should be pursued experimentally.

The three major compositional types are not all found in each of the three major textural occurrences. Only the following have been found:

<table>
<thead>
<tr>
<th></th>
<th>High-Fe</th>
<th>High-K</th>
<th>Low-K</th>
</tr>
</thead>
<tbody>
<tr>
<td>Melt inclusions in:</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ilmenite</td>
<td>×</td>
<td>×</td>
<td>×</td>
</tr>
<tr>
<td>Pyroxene</td>
<td></td>
<td>×</td>
<td></td>
</tr>
<tr>
<td>Plagioclase</td>
<td>×</td>
<td>×</td>
<td></td>
</tr>
<tr>
<td>Pyroxferroite</td>
<td>×</td>
<td>×</td>
<td></td>
</tr>
<tr>
<td>Cristobalite</td>
<td>×</td>
<td>×</td>
<td></td>
</tr>
<tr>
<td>Mesostasis (interstitial inclusions)</td>
<td>×</td>
<td>×</td>
<td></td>
</tr>
<tr>
<td>Clasts in breccias</td>
<td>×</td>
<td></td>
<td>×</td>
</tr>
</tbody>
</table>

The absence of recognizable high-Fe melt as inclusions in pyroxene and pyroxferroite is expectable, since high-Fe melt crystallizes readily when in contact with such phases. Similarly, the post-shock metamorphism that has affected many of the breccias would also cause any clasts of high-Fe glass to crystallize and hence lose their identity. The occurrence of the low-K melts solely as inclusions in ilmenite is another perplexing, but possibly significant aspect to the problem of the nature and origin of this peculiar melt. Rare intergrowths of crystalline plagioclase and silica have been described in some
Compositional variation in late-stage differentiates in mare lavas

Apollo 17 breccias (Ryder et al., 1975) that might represent the crystallized equivalents of low-K melts. The most notable occurrence of the high-K melt composition as clasts is the Apollo 14 breccias (Roedder and Weiblen, 1972, their Table 6), but they are also common in some Apollo 17 breccias, particularly 72255 (Stoesser et al., 1974; Ryder et al., 1975).

Bulk chemical composition

In Table 2, we list the average chemical composition of the three major chemical types of late-stage differentiates from Apollo 15 and 17 samples. These specific samples were used, rather than a grand average of all analyses, as there are significant differences between the data from various missions (e.g., Roedder and Weiblen, 1972b, p. 281). The averages in Table 2 are based on our previously published analyses (also included in Table 1). Analyses from individual rock groups, and particularly from individual inclusions, will differ from these averages, as detailed below, but most differences in averages are not large.

GROUPING AND RECOGNITION OF ROCK TYPES ON THE BASIS OF LATE-STAGE INCLUSION ANALYSES

Many classifications of mare basalts have been made on the basis of bulk chemistry, the most thoroughly documented being that of Papke et al. (1976). The differences in composition between these types, though analytically significant, are small. The late-stage melt inclusions, however, represent a sample of the residue after crystallization of \( \approx 95\% \) of the original magma (Roedder and Weiblen, 1970). For those elements that are concentrated in the late-stage melts, absolute differences are enhanced. Thus inclusions from different magmas should show large relative differences, even though the differences in the original magma compositions were small. We have found that this is indeed true. Differences in the composition of the melt inclusions in the various samples and rock types are far larger than the expected analytical errors and are sufficient to permit recognition of some rock groups, at least, even as small fragments in breccias. This has the added advantage of requiring only nondestructive electron microprobe analyses, whereas bulk chemistry for either major or minor elements on a crystalline rock requires the destruction of an appreciable amount of sample, and hence would not be suitable for small fragments in breccias.

In order to recognize relationships between the many variables represented in Table 1 (sample number, rock type, mission, host mineral, stage of differentiation, and eight major elements), a variety of computer plots was made. These are far too numerous and large to be reproduced here, so we present only a few of them. The relationships are much more easily visualized by the reader, and can be presented much more briefly, when described in terms of how the points fall on a diagram, so we will use this artifice for presenting the salient points from other diagrams not reproduced here. Copies of these various diagrams are available from the authors.
Table 2. Average chemical composition of the three major types of late-stage differentiates. Data from Table 1. Number of analyses averaged given in parentheses.

<table>
<thead>
<tr>
<th></th>
<th>Apollo 17 high K (32)</th>
<th>Apollo 15 high Fe (8)</th>
<th>Apollo 17 low K (57)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Max.</td>
<td>Min.</td>
<td>Mean</td>
</tr>
<tr>
<td>SiO₂</td>
<td>80.3</td>
<td>67.8</td>
<td>76.31</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>17.1</td>
<td>8.34</td>
<td>12.10</td>
</tr>
<tr>
<td>FeO</td>
<td>8.46</td>
<td>0.77</td>
<td>2.20</td>
</tr>
<tr>
<td>MgO</td>
<td>0.66</td>
<td>&lt;0.05</td>
<td>0.23</td>
</tr>
<tr>
<td>CaO</td>
<td>5.80</td>
<td>&lt;0.05</td>
<td>1.80</td>
</tr>
<tr>
<td>Na₂O</td>
<td>1.43</td>
<td>&lt;0.05</td>
<td>0.41</td>
</tr>
<tr>
<td>K₂O</td>
<td>10.30</td>
<td>3.98</td>
<td>5.95</td>
</tr>
<tr>
<td>TiO₂</td>
<td>2.19</td>
<td>0.05</td>
<td>0.68</td>
</tr>
<tr>
<td>P₂O₅</td>
<td>0.65</td>
<td>&lt;0.05</td>
<td>0.05</td>
</tr>
<tr>
<td>MnO</td>
<td>0.11</td>
<td>&lt;0.05</td>
<td>0.01</td>
</tr>
<tr>
<td>BaO</td>
<td>2.11</td>
<td>&lt;0.05</td>
<td>0.13</td>
</tr>
<tr>
<td>Cr₂O₃</td>
<td>0.05</td>
<td>&lt;0.05</td>
<td>&lt;0.01</td>
</tr>
<tr>
<td><strong>Totals</strong></td>
<td>101.75</td>
<td>97.68</td>
<td>99.86</td>
</tr>
</tbody>
</table>
In Figs. 5 and 6, we illustrate a few of the differences between groups. In Fig. 5, for example, note that the points for 10062 are offset from those for 10044 and 10047. This difference is important, as 10044, 10047, and 10062 are all classified as Apollo 11 low-K basalts (Papke et al., 1976). Although the grain size of 10062 is smaller than that of the other two, so the cooling rates were probably not the same, there are also differences in the bulk chemistry of these rocks, particularly in Si/Al and Fe/Mg. In Fig. 6 note that the average for Apollo 14 granitic clasts is distinct from the “granitic” differentiates found in the Apollo VHTi basalts. Similar distinctions were found between the “fields” of 10047 and all Apollo 15 high-K inclusions on plots of K₂O vs. SiO₂ and FeO/(FeO + MgO) vs. SiO₂. Very few points have been excluded from these plots on the basis of the factors discussed above, but this exclusion results in a tighter clustering of points in each group.

A strong clustering of points is found on many of the other plots. Thus, on a plot of FeO/(FeO + MgO) vs. SiO₂ for all high-K inclusions from all missions (304 analyses, including Luna 16 and 20), more than two-thirds of the points fell in an area that constituted only about 5% of the area necessary to include all points. The points representing averages for individual rock samples or rock groups show a much smaller compositional range than the individual analyses, of course, but even these averages reveal analytically significant differences. As an example, on Fig. 7 we plot K₂O vs. SiO₂ for the high-K inclusion averages (18) for various rock groups; these range from 5–9% K₂O and from 73 to 78% SiO₂. Similarly, in the various groups of low-K inclusions, although the variations in

![Fig. 5](image1.png)

![Fig. 6](image2.png)

Figs. 5–6. Plots of the composition of silicate melt inclusions in ilmenite from various samples, grouped as indicated for comparison purposes. The number of points plotted is given in parentheses. Data from Table 1. The lines outlining the “fields” are drawn solely as a visual aid.
K$_2$O and FeO/(FeO + MgO) are small, the CaO ranges from 5 to 8% and the SiO$_2$ from 74 to 81%. Yet each of the averages represents very sizeable numbers of samples.

We have no ready explanation for many features in such plots. Thus, the inclusions in some individual rock samples show significant subgroups. Twenty-three high-K melt inclusions were analyzed from slide 10047,11. Of these, 16 fall in a relatively tight cluster on all plots (e.g., Fig. 6), and the remaining seven, although essentially identical with the 16 in terms of occurrence and Si, Ca, and Na, are markedly different in other respects and fall in two separate subgroups. Such groups are too tightly clustered to have the differences assigned to simple random sampling and analytical errors; ad hoc explanations of these differences can usually be found, such as local equilibrium reflecting the crystallization of another phase nearby, but these explanations are not very satisfactory.

One characteristic trend was found—a small but significant negative correlation between K$_2$O and SiO$_2$ within various groups of samples. This is apparent in a plot of all high-K averages (Fig. 7), where the 18 points delineate a rough line with a negative slope. We believe this is only in part a result of the requirement of a negative correlation between any two major constituents, if all others are held constant. The points outlining such trends, which are based on data on the residual melts in a series of independent, different magmas, do not necessarily imply derivation of one inclusion composition from another, as might be assumed for a similar trend observed in a suite from a single sample.

**Bimodality of Potassium Values**

Roedder and Weiblen (1975) showed that 87 melt inclusions either in or interstitial to late ilmenite all had $\sim$76% SiO$_2$, but they had a very strongly

![Fig. 7. Plot of the average compositions of high-K silicate melt inclusions (both in ilmenite and interstitial) from various individual samples or missions (data from Table 1). Individual points represent averages of 3–35 analyses, with an average of 11 analyses.](image-url)
bimodal K₂O distribution: one group averaged 0.03% K₂O, whereas the other averaged 6.4%, and only three were in the range 0.15–4.05% K₂O. A new plot of K₂O for the 309 such inclusions in Table 1 (Fig. 8) shows that although there are now analyses in almost every K₂O concentration class interval, the bimodality is statistically secure, one group averaging ~6% and the other near 0% K₂O. Note that the average of all 309 analyses falls at ~2.8% K₂O, at the center of the low point on the histogram. We still do not have a satisfactory explanation for the origin of the extremely low-K melts. We have previously listed 13 suggested mechanisms (Roedder and Weiblen, 1975), but found them all unsatisfactory to some degree.

**Delineation of Differentiation Trends**

We have found a plot of K₂O vs. SiO₂ to be particularly illustrative of the compositional range of late-stage differentiates. In Fig. 9, we show K₂O vs. SiO₂ for our data on all types of inclusions, and in Fig. 10 we show an expansion of the low-K part of Fig. 9. Also plotted are the residual melts found by Rutherford et al. (1974) in laboratory fractionation studies on a synthetic composition.

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**Fig. 8.** Frequency diagram for K₂O contents of 309 inclusions (interstitial and in ilmenite) in Apollo 11, 12, 15, and 17 basalts. The data from individual missions differ significantly (Table 1), but all yield similar bimodal K₂O distributions.
duplicating 70017, and those found in some Hawaiian samples (taken from Weiblen and Roedder, 1976, p. 1455; Wright and Weiblen, 1968; Wright et al., 1976).

Several features are evident on these two plots. *First*, although some ilmenite trapped early melts that were just as low in SiO$_2$ and K$_2$O as those trapped in olivine, and hence some of the ilmenite crystallized early, no inclusions with very high SiO$_2$ and only three with high K$_2$O were trapped in olivine. This suggests that crystallization of olivine generally ceased before such melts were formed. A more detailed discussion of the early differentiation trends shown on the lower left side of Figs. 9 and 10 is given in a companion paper by Weiblen (1977). It is also evident that there is no real difference in the compositional range for inclusions in ilmenite vs. those that are interstitial, except in the very high-K range. *Second*, a significant gap at 2–3% K$_2$O may possibly indicate liquid immiscibility, although this gap occurs over a range of SiO$_2$ values. All points from Rutherford et al. (1974) above 1.9% K$_2$O represent analyses of fine-grained mixtures of two immiscible melts (P. C. Hess, pers. comm., 1977). *Third*, if immiscibility involves simply the splitting of a liquid to form high-Fe and high-Si melts without simultaneous crystallization, then the parent liquid just before splitting has to lie along a line connecting the two compositions just after the liquid has split. As there are no analyses with 2–3% K$_2$O near the line connecting these two, either we have fortuitously failed to sample the liquid just before splitting, which seems unlikely in view of the number and SiO$_2$ range of our analyses, or some other mechanism must be involved. Since the melt probably arrived at the field of immiscibility as a consequence of crystallization, it is probable that additional separation of the immiscible melts also entails formation of a third phase, such as SiO$_2$, so that the trends would not be explicit on Figs. 9 and 10 without knowledge of the relative amounts of phases. *Fourth*, FeO/(FeO + MgO) generally increases with degree of crystallization. We have examined this ratio for the analyses plotted on Figs. 9 and 10 and find that it increases with SiO$_2$ and that on a plot vs. K$_2$O, low values for the ratio occur only with low values of K$_2$O. At high-silica values the ratio is variable; this is probably due to the fact that since most of the magnesia has been used up before the melt reaches even 60% SiO$_2$, formation of relatively minor amounts of crystals may effect large changes in the ratio. *Fifth*, we had

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**Fig. 9.** K$_2$O vs. SiO$_2$ for all 826 selected melt inclusion analyses: octagons with vertical line = inclusions in olivine; triangles = inclusions in spinel, plagioclase, and pyroxene; (+) = inclusions in ilmenite; (×) = re-entrant and interstitial inclusions, plus ten K-spars (bracketed; these contain significant barium). For comparison, we include data on the residual melts obtained after partial crystallization in experimental runs on composition 70017 (solid circles; Rutherford et al., 1974); a differentiated series of samples from Hawaii, ending in a granophyre (small open circles; Weiblen and Roedder, 1976, p. 1455; Wright and Weiblen, 1968; Wright et al., 1976); and average high-Fe and high-K inclusions (Apollo 11; large circles). Only the top edges of the symbols for points along the abscissa show in this plot; the portion from 0–2% K$_2$O is expanded in Fig. 10. A mixing line between stoichiometric K-spar and SiO$_2$ (squares) is indicated.
Fig. 10. K₂O vs. SiO₂ for all inclusion analyses with less than 2% K₂O (expanded from bottom edge of Fig. 9). Symbols as given in Fig. 9.
hoped that such plots would show a relatively linear differentiation trend, as would be required if all the inclusion samples were glassy and cogenetic. The spread of the data on Figs. 9 and 10 and similar plots presumably reflects the combination of different trends for the various original magma compositions, liquid immiscibility, local variations in crystal-liquid equilibria at the time of trapping, sample heterogeneity from crystallization, and analytical errors. The broad scatter in the high-K range and the extremely high SiO$_2$ values are probably due mainly to inhomogeneity from K-spar and silica minerals in the inclusions; these minerals cannot be easily distinguished from high-Si glass in reflected light or by cathodoluminescence in this kind of occurrence. As the high-Si, high-K inclusions contain only 70–90% normative K-spar plus silica, they do not fall on the K-spar/silica mixing line (Fig. 9) but are dispersed more or less parallel to it. Last, the trend from Rutherford et al. (1974) and from the Hawaiian samples fits a curved line between the bulk rock composition of mare basalts (which would plot in the lower left corner of Fig. 9) and our Apollo 11 high-K melt average. In theory, these high-K compositions can be derived from the bulk rock by crystallizing olivine, ilmenite, and pyroxene. Most inclusion compositions fall on the higher SiO$_2$, lower K$_2$O side of this line, however. As the parent magmas have very low K$_2$O, the crystals must have even lower K$_2$O to yield a net K$_2$O enrichment in the melt. Thus, 97% crystallization with 0% K$_2$O will leave 3% of melt enriched 33-fold in K$_2$O (e.g., from 0.04 to 1.32%). Rutherford et al. (1974) assumed 98.3% crystallization of K$_2$O-free phases (including plagioclase) to obtain the 3% K$_2$O melts. Many lunar plagioclases have enough K$_2$O to result in a negative slope on Fig. 9. We found ~20% modal plagioclase in 70017; this would have to average 0.25% K$_2$O for the calculated residual liquids to show 0% K$_2$O enrichment. Although analyses of plagioclase from a similar basalt, 75083 (Papike et al., 1976) show 0.02–0.28% K$_2$O, it seems highly unlikely that the crystallization of plagioclase could keep the residual liquid depleted in K$_2$O to the extent shown by many points on Figs. 9 and 10. In fact, in 70035, we found that even plagioclase crystallizing from a high-K melt contained only 0.35% K$_2$O in the outermost zone. Thus the paradox remains: all available data point to K$_2$O enrichment in the residual melts, yet many glassy inclusions in ilmenite have low K$_2$O and high SiO$_2$, and no mechanism seems adequate to explain this fact (Roedder and Weiblen, 1975).

CONCLUSIONS

Many of the problems in interpretation and much of the scatter found in the composition of silicate melt inclusions in lunar samples can be attributed to the incomplete nature of the record and to crystallization after trapping. These sources of difficulty were minimized by the selection of appropriate sample material. The resulting data bank: (1) makes possible the grouping and recognition of some rock types, even in breccia fragments, on the basis of analysis of melt inclusions; (2) verifies the still-inexplicable bimodal distribution of K$_2$O values for high-Si melt inclusions in ilmenite; and (3) permits delineation of some
differentiation trends, and, in particular, suggests the intervention of silicate liquid immiscibility when these residual melts have reached about 2% K₂O.

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