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Petrologic data from experimental studies on crystallized silicate melt and other inclusions in lunar and Hawaiian olivine

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Abstract

Inclusions trapped in olivine of lunar mare and Hawaiian lava lake basalts have yielded data on the following: (1) composition of the melt at the time of olivine crystallization, except for subsequent crystallization of olivine on the walls (from probe analyses on natural or laboratory homogenized glasses); (2) amount and nature of immiscible sulfide melt, and the stage at which it separated (from simple microscopy); (3) equilibrium temperatures and sequence of melting, the nature of the phases other than olivine crystallized *within* the inclusions during cooling, and a crude measure of the solidus temperature of the trapped fluids (from phase-disappearance heating experiments on crystallized inclusions); (4) relative cooling rates of different flows and some constraints on absolute cooling rates (from simple microscopy and annealing experiments); (5) possible distinctions between phenocrysts and exotic xenocrysts (from the composition of inclusions); (6) presence of a vapor phase at the time of growth of the olivine and some data on its composition (from crushing-stage studies); and (7) minimum pressures at the time of olivine growth (from gas inclusions).

Limitations are imposed by the suitability of the available inclusions, by the implicit assumptions, particularly as to the representativeness of the sample, and by the experimental techniques. Not all types of data can be obtained on any given sample, and several important questions and conflicts remain unanswered. In spite of these shortcomings, some of these data are believed to be valid, and some cannot be obtained otherwise.

Introduction

Skeletal crystallization of olivine in both lunar and terrestrial basalts and in ultramafic nodules frequently causes trapping of primary melt inclusions, which are thus isolated from the main mass of the melt. When trapped, they contain a full complement of volatile materials and generally are unaffected by later minor deuteric alteration or oxidation of the host olivine.

As even a large inclusion (~200 μm in diameter) is actually a very small system that may be completely free of heterogeneous nucleation centers, metastable equilibria from failure to nucleate phases are commonly found. Whether or not other phases nucleate in the trapped melt as cooling takes place, further crystallization of olivine on the walls of the inclusion will change the composition of the remaining melt along an extension of the olivine control line (*i.e.*, a line connecting compositions differing only due to crystallization of olivine). The natural cooling rates for some inclusions have been sufficiently slow, however, to permit nucleation and growth of one or more new *daughter* phases other than olivine in the olivine-saturated melt. The most common are: *"vapor"* bubble (2–3 vol. percent, formed by differential shrinkage of crystal host and melt), an immiscible *sulfide* melt (~0.1–0.5 vol. percent; Fig. 8), *pyroxene* (either as a few coarse crystals or as a feathery mat), *plagioclase,* and *ilmenite.* The last two frequently occur as parallel flat plates, epitaxially oriented on the olivine substrate walls (Roedder and Weiblen, 1970).

In addition to the daughter phases, other phases that were present at the time of trapping may be enclosed with the melt. The most common of these *primary* phases are spinel (the enclosing of which frequently causes the trapping of melt as well), globules of high- to low-pressure CO_2 *gas,* verified by the use of the crushing stage (*i.e.*, incipient vesiculation; Figs. 4, 6, and 14; Roedder, 1965), and immiscible *sulfide melt* (*cf.* Fig. 9; Roedder and Weiblen, 1970, p. 810).

Silicate-melt inclusions in olivine, now consisting of six phases, including epitaxial daughter crystals of plagioclase and ilmenite, were first reported from the lunar mare basalts (Roedder and Weiblen, 1970) and were then found to be common and obvious in samples from some Kilauean lava lakes (Figs. 1–3; Roedder and Weiblen, 1971). These crystallized inclusions, and to a lesser degree the other types, provide a variety of useful data on the growth environment and thermal history of the host olivines, as well as compositional data on the magma involved. This paper mainly concerns three types of data that can be obtained from inclusions: relative cooling rates, equilibrium crystallization sequence, and constraints on the absolute cooling rates.

Relative cooling rates determined from simple microscopy of the natural inclusion phase assemblage

Useful qualitative data on relative cooling rates of lavas can be obtained from simple microscopy. This can be done by classifying the inclusions into five "crystallinity" categories, depending upon the nature and amount of nucleation and growth of daughter phases (Roedder and Weiblen, 1971, p. 512). These categories are necessarily rather crude but do permit at least semiquantitative comparisons. The two extreme categories are *all glass,* at one end, and *epitaxial ilmenite and plagioclase* (± pyroxene) at the other. (Nucleation and growth of new fluid phases—vapor and sulfide melt—are ignored in this classification but probably follow a pattern similar to that of the crystalline phases.) Intermediate categories between the two extremes are inexact and arbitrary. They consist of *few random crystals, many random crystals,* and *epitaxial ilmenite only* (± pyroxene), in presumed order of slower cooling and increasing "crystallinity."

The nucleation and growth of such new phases is a complex function of many variables, the most important of which are inclusion volume, cooling rate, and composition. Nucleation in these little systems, both heterogeneous and homogeneous, is obviously a function of the *volume* of the system. Thus there will be only $\sim 10^{10}$ atoms present of each element occurring in the 1 weight percent range in the smaller inclusions studied here, so the probabilities of overcoming the barriers against homogeneous nucleation of a phase involving these elements will certainly be affected. Similarly, the probabilities of trapping, in any given inclusion, a particle of some extraneous phase that might act as a nucleus for heterogeneous nucleation will also increase directly with inclusion size. The effects of inclusion volume can be effectively eliminated by comparing inclusions of equal volume.

Both nucleation and growth of new phases will also be a function of the *cooling rate* through specified optimum ranges. These ranges will generally be different for nucleation and for growth of a given phase, and probably different for each phase. Furthermore, it has been found that the previous thermal history of glasses may seriously affect their devitrification rates, so basaltic glass may also be similarly affected.

In addition to volume and cooling rate, the *composition,* particularly the content of volatiles, of the original melt that was trapped must obviously have an effect. Anderson (1974a, p. 1488) has shown that the rather large amounts of H_2O present in some melt inclusions in olivine apparently could leak or diffuse out of the inclusions within hours in laboratory experiments. Except for sulfur, the volatile materials (particularly H_2O) present in the basaltic lavas examined in the present study, both lunar and terrestrial, are probably very low. The gross composition of the melt, particularly the silica content, is also of great importance in nucleation and crystallization rates. Although the various types of basalts studied are far from identical in composition, I believe that the differences in nucleation behavior due to composition are probably of second-order importance.

The results of examination of inclusions in olivine phenocrysts from eight basalts are shown in Figure 20. A ninth sample, 201 inclusions from slides 34 and 253 of lunar sample 15555, showed a histogram very similar to that of sample 12036 in Figure 20. The growth history of the Hawaiian (as well as the lunar) phenocrysts may not be simple. Some show clear evidence of two stages of growth, under changing conditions (Figs. 4 and 14). As the early cores constitute only a very small percentage of the volume of the olivine, and as no differences were recognized in the inclusions, this two-stage growth probably does not affect the conclusions reached here. When inclusions of equal volume (*i.e.*, a given size range) are compared in the four lunar samples in Figure 20, the relative cooling rates can be assumed to increase from left to right. The relative cooling rates of the four terrestrial samples are not as clear. The effects of volume on the probability of nucleation are obvious in Figure 20; except for the sample from Kilauea Iki ("KI"), the degree of "crystallinity" increases rather regularly as inclusion size increases.

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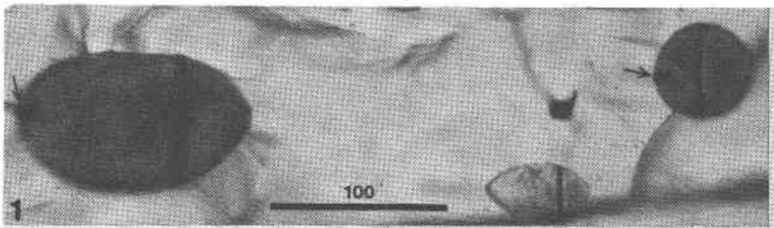

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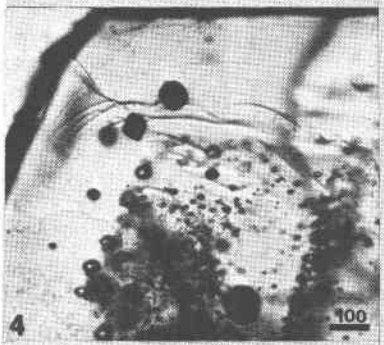

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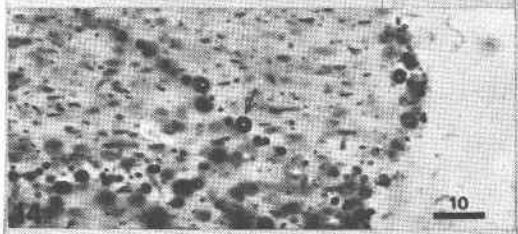

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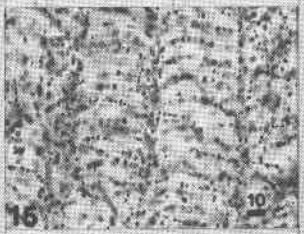

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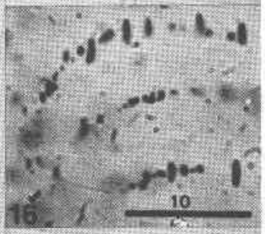

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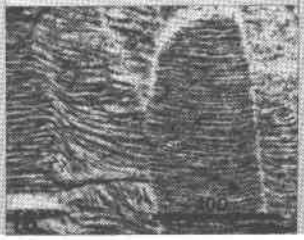

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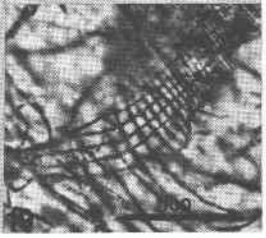

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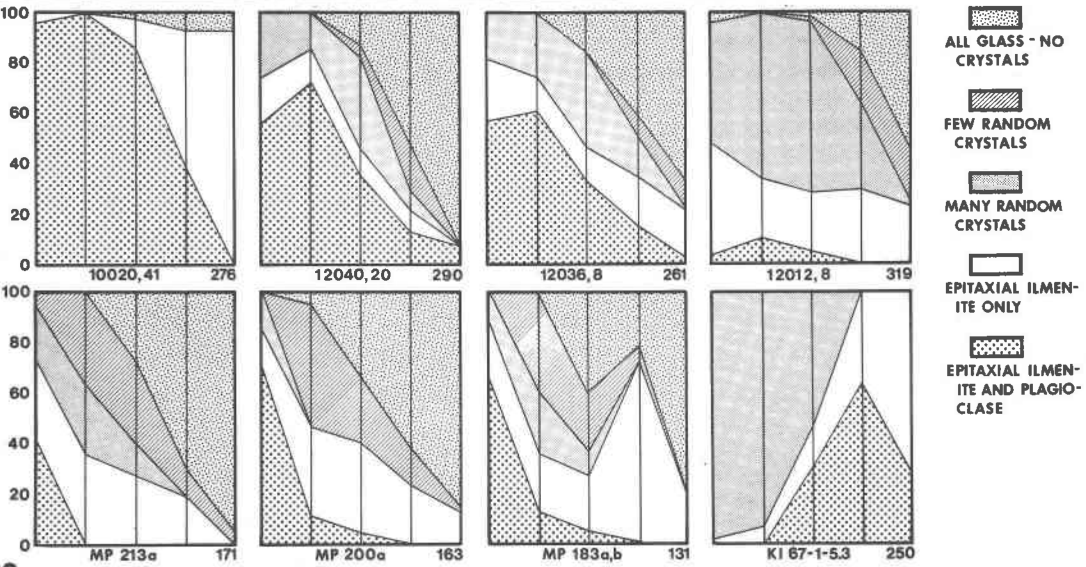

Notes. All photomicrographs are of olivine, were taken in transmitted plain light, and have scale bar dimensions in μm. "MP" samples are from the prehistoric Makaopuhi lava lake (subtract 50 from the number to get depth in lake in feet); "KI" samples are from the Kilauea Iki lava lake.

FIG. 1. Three of six large inclusions, all in same phenocryst, containing epitaxial ilmenite (black bars) and plagioclase (light zones adjacent and parallel to the ilmenite) in feathery pyroxene (and plagioclase?). Two sulfide globules are visible (arrows) (MP 200).

FIGS. 2 AND 3. Most common range of ratio of epitaxial ilmenite (black bars) to plagioclase (clear). (MP 200a).

FIG. 4. KI-67-1. Olivine phenocryst showing inclusion-rich core (lower right), outlined by a row of gas inclusions (bright centers), and covered by relatively inclusion-free overgrowth.

FIG. 5. KI-67-2-94.7 (ft.) depth. Symplectitic intergrowth.

FIG. 6. MP 213a. Primary gas inclusion containing a small amount of glass (bottom).

FIG. 7. KI-67-1-7.2 (ft.) depth. Small inclusion (9 μm), containing epitaxial ilmenite and two epitaxial plagioclase crystals (arrows).

FIGS. 8 AND 9. Adjacent tiny inclusions in sample MP 200a. Fig. 8 shows typical immiscible sulfide globule, believed to be a true daughter phase, whereas the globule in Fig. 9 probably represents trapping of a primary sulfide globule.

FIGS. 10 AND 11. MP 200a. Thick and thin epitaxial ilmenite plates (arrows) in tiny inclusions.

FIG. 12. Melt inclusion in MP 205, originally 6-phase, after homogenization at 1230°C for 1 hour, cooling from 1230° to 1120°C over 5.5 hours, then 70 hours at 1120°C; contains sulfide, glass, vapor, and a pyroxene crystal.

FIG. 13. KI-67-1-23.2 (ft. depth). Inclusions showing sulfide, glass, and coarsely crystalline pyroxene.

FIG. 14. Part of phenocryst from MP 213a showing on left the crystal core containing exsolved opaque phase (thin black bars), and planes of pseudo-secondary gas inclusions (CO_2?; arrow), all covered with new, optically continuous inclusion-free overgrowth (right).

FIGS. 15 AND 16. General view and detail of decorated dislocations(?) in olivine of MP 213. The opaque plates are oriented parallel to an extinction direction of the host olivine and have a reflectivity about equal to that of ilmenite.

FIGS. 17–19. Decorated dislocations(?) in olivine from KI-67-1-60.2 (ft. depth).

FIG. 20. Histograms showing abundance of various types *vs.* maximum dimensions for inclusions in olivines from lunar and Hamaximum dimensions for inclusions in olivines from lunar and Hawaiian lavas. The size brackets for the five sample subsets in each histogram, from left to right, are as follows: >40, 20–40, 10–20, 5–10, and <5 μm. The number of inclusions measured in each sample is given at lower right of each graph. The first three histograms in upper row are in large part adapted from Roedder and Weiblen, 1971. Kilauea Iki sample (lower right) from 1967 borehole 1, at 5.3 ft. depth. Pyroxene may be present in either of the two epitaxial categories.

My interpretation is that cooling through the range of nucleation of ilmenite and plagioclase was sufficiently slow in most of these samples so that, in the larger inclusions, only one crystal of each phase formed and grew epitaxially on the olivine substrate. Cooling was too fast for crystal nucleation in most of the smaller inclusions; note, however, that epitaxial ilmenite did nucleate even in some very small inclusions (Figs. 7 and 9–11). An interesting divergence from these general trends is shown by the abrupt rise in percentage of inclusions containing epitaxial ilmenite only in the small size range in sample MP 183. This might be passed off as merely statistically expectable scatter in a small sample, but this particular histogram data class consisted of 50 inclusions,[1] so the large percentage is probably valid, though of unknown origin. The Kilauea Iki sample KI-67-1-5.3 (ft.) differs drastically from the other seven in that it has a "reversed" distribution.[2] This may indicate a two-stage cooling history: first, a period of cooling in the range of temperature favoring optimum nucleation of pyroxene (and plagioclase?), resulting in abundant random crystallization in the larger inclusions (some containing interstitial ilmenite), but during which the smaller inclusions stay glassy. Second, an extended period of cooling in a presumably lower range of temperature causing nucleation of epitaxial ilmenite and plagioclase but not pyroxene in the remaining small inclusions. Although this explanation for the reversed distribution is special for this case alone, such a cooling history is certainly feasible in a lava lake which had repeated foundering of crust and repeated drainbacks (Richter *et al.*, 1970).

Once a lava lake is crusted over, the subsequent cooling history of most samples should be closely related to their depth in the lake. However, in samples from the prehistoric Makaopuhi lava lake (see Moore and Evans, 1967; Evans and Moore, 1968, for detailed petrology), I found both wholly glass inclusions and even small inclusions containing epitaxial ilmenite and plagioclase *throughout* the entire series, from a sample taken 3 feet below the top to one taken in the chill zone at the very bottom (Moore and Evans, 1967; their samples MP 53 and MP 275). If we assume, as a rough first approximation, that 1050°–1000°C is the critical range for nucleation of epitaxial ilmenite and plagioclase, the calculated cooling curves of Jaeger (Evans and Moore, 1968; their Fig. 2) show that the three Makaopuhi samples in Figure 20 (MP–183, -200, and -213, from depths of 133, 150, and 163 feet, respectively), took about 20 years to cool to 1050°C, and 10 more years to cool to 1000°C. Cooling rates on the bottom chill zone are difficult to estimate, but the rate for the sample from a depth of 3 feet (MP 53), may be approximated from data on the 1965 Makaopuhi lava lake (T. L. Wright, in preparation). In this lake a sample from a depth of 3 feet would have cooled to 1050°C in about 8 days and would have reached 1000°C 2 days later. Why these vastly different cooling histories should give similar inclusion results is an unanswered question. An added complication here, of unknown significance, is the fact that after the lava in the prehistoric lake had cooled to the point of forming columnar joints throughout (~20 years), a new series of flows (~50 feet thick) was extruded over the top, and hence may have baked the uppermost samples a second time. Unfortunately, most of the available slides do not have enough olivine to permit the statistically significant sampling shown in Figure 20. The broad distribution of inclusions with epitaxial daughters might suggest that they were present on eruption, but this possibility is precluded by the heating experiments described below, and by the fact that such inclusions have not been found in any air-quenched sample of recent lavas.

Some degree of "calibration" of these data is possible by using U.S. Geological Survey drillhole samples from the crust of the new 1965 Makaopuhi lava lake. These samples had known time-temperature histories, and some were quenched from known temperatures (T. L. Wright, in preparation). Of the 36 samples examined from various boreholes in the 1965 Makaopuhi lake, only seven show epitaxial daughter crystals. Six of these are from hole 13, and were quenched from temperatures of 380°–500°C (minimum) to 1020°C (maximum), 80 days after eruption. The other was in hole 11, sampled at 725°–795°C after 67 days. These samples would have passed through the range 1050° to 1000°C in about 20 days. All but one of the other samples examined, in which I found *no* epitaxial crystals, had been quenched from temperatures above 1020°C, although some of these were sampled >80 days after eruption. The one exception was taken from very near the surface and hence was sampled at near-surface temperature but presumably cooled very rapidly. Epitaxial crystals were also present in samples from a wide range of

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[1] The average histogram bar in Figure 20 is based on 47 inclusions, and only three bars are based on less than 20 inclusions.

[2] This divergent distribution can hardly be attributed to statistical fluctuations, as the numbers of inclusions in the five classes in this diagram, from left to right, are 43, 55, 61, 54, and 37.

depths in the 1967 boreholes in the Kilauea Ike lava lake (Fig. 7). From this, it is evident that the epitaxial daughter crystals can but do not always form during cooling through the range 1020°–~800°C at 2°–3°C/day. The available data do not permit any closer constraints on either range or rate.

Equilibrium crystallization sequence determined from heating experiments on naturally crystallized inclusions

Although natural nucleation of various crystalline phases on cooling did not necessarily occur in the stable phase-equilibrium order, once the correct phases are present, progressive heating of such crystallized inclusions can give valid data on the equilibrium crystallization sequence for the olivine-saturated melt (Roedder and Weiblen, 1970).[3] As oil-immersion objectives are needed for phase identification, such studies require a sequential cycle of heating, quenching, and observation, repeated at increasingly higher temperatures. Selected olivine grains (~0.5–1.0 mm) are held in individual unsealed iron foil envelopes, which are held in turn in evacuated fused-silica tubes. Losses can be frustratingly high from cracking and leaking during the runs, sticking due to melting of adhering grains of other phases, grains becoming opqaue due to surface etching or films, or simple ineptness with the tweezers on grain transfer, but some individual grains have been successfully put through many such heating cycles without loss. At any stage, the inclusion may be sectioned for microprobe analyses, but this terminates the runs on that inclusion, as the olivine "bottle" is then open. Runs of a few hours at temperature seem to be adequate to achieve equilibrium in phase-disappearance runs.

Some earlier heating runs on samples of olivine from the prehistoric Makaopuhi lavas (Roedder and Weiblen, 1971) have been extended and are presented here, although still preliminary: apparent solidus 1040° ± 10°C; plagioclase out 1065° ± 25°C; ilmenite out 1080° ± 10°C; pyroxene out 1135° ± 5°C; vapor bubble out and probable trapping temperature 1220° ± 10°C. The sulfide daughter globule dissolves at about 1220°C, but this temperature is inexact because of the presence of primary immiscible sulfide globules in the lavas at the time of trapping (*cf.* Figs. 8 and 9). If such a globule becomes trapped, the inclusion will obviously not be representative of the silicate melt composition.

A discrepancy exists between these data on *olivine-saturated* melts and the measured temperatures for the appearance of pyroxene and plagioclase in the 1965 Makaopuhi lavas (Wright and Weiblen, 1967). In these samples, they found that clinopyroxene started crystallizing at 1185° ± 10°C, plagioclase at 1175° ± 10°C, and ilmenite at 1070° ± 10°C. Although the 1965 lavas have a bulk composition for the whole lake that is almost identical with the prehistoric ones, they were *quartz-normative* at this stage, because of sinking of olivine; it is not known whether this difference is adequate to explain the 50°C discrepancy in the temperature of crystallization of pyroxene and ≥75°C for plagioclase.

Constraints on the absolute cooling rates from laboratory nucleation studies on glassy inclusions

If inclusions are glassy when they are first found, but nucleate crystals during a given laboratory cooling cycle, the natural cooling cycle must have been faster than the laboratory one. The possibility that the natural cooling cycle resulted in glass annealing that affected the nucleation can be eliminated by the use of laboratory-homogenized inclusions in samples that also contain glassy inclusions. Inclusions in olivine samples from the prehistoric Makaopuhi lava lake were homogenized at 1230°C, and then cooled from 1230° to 1120° over 5.5 hours. On examination, all inclusions but one had glass, vapor, and sulfide. The exception has also nucleated a few coarse pyroxene crystals (similar to those shown in Fig. 13). These same samples were then held at 1120° for 70 hours, but no further change was noted except nucleation of a single pyroxene crystal in another inclusion (Fig. 12). These data place some constraints on the cooling rates needed to nucleate coarse pyroxene crystals, such as are found in some of the natural inclusions (Fig. 13) and are compatible with the phase-disappearance data (pyroxene out at 1135 ± 5°C), but none of the laboratory runs so far have yielded the feathery pyroxene that is more common in the naturally crystallized inclusions. Epitaxial ilmenite crystals grew in glassy homogenized inclusions from the prehistoric Makaopuhi samples during a cooling from 1120° to 1020°C over an 11-day period (Roedder and Weiblen, 1971, p. 510).

One possible source of previously untapped thermal data is the exsolution of trace constituents in

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[3] This procedure is equivalent to the "seeding" of laboratory phase-equilibrium runs with specific phases, but the inclusions are 3 to >6 orders of magnitude smaller than normal 10^{-3} g laboratory charges.

these same olivines. This exsolution either decorates dislocation arrays (Figs. 14–19) or forms much larger but still microsymplectitic intergrowths. These two types of features were first observed in lunar olivines (Roedder and Weiblen, 1970, 1971), and correspond to symplectite types F and A of Bell *et al.* (1975). The sporadic occurrence probably results from compositional variations in the olivine from a two-stage origin (*e.g.*, crystal growth before and after eruption; Figs. 4 and 14), rather than from disequilibrium. As the decorated dislocations in the lunar samples became much more prominent after laboratory heat treatment (Roedder and Weiblen, 1970, Fig. 9, p. 807), rate studies on this exsolution should be compared with and should be compatible with the melt inclusion data.

Other types of data obtainable from inclusions

In addition to the above three general types of data, glassy inclusions or laboratory-homogenized ones can be analyzed by electron microprobe to obtain the composition of the magma from which the olivine crystallized (after a correction for the composition change due to further crystallization of olivine in the walls; Roedder and Weiblen, 1970, p. 810; Anderson, 1974b). Such analyses should also permit distinction between phenocrysts and exotic xenocrysts (Anderson, 1971). The presence of inclusions of gas in some of these samples (Figs. 4, 6, and 14) can be used to place some limits on the pressure on the system at the time of trapping, under the assumption that the inclusions probably contain mainly CO_2 (Roedder, 1965). As no visible liquid CO_2 formed in these on cooling, but as the inclusions now contain gas under ~1 atm pressure, these inclusions were probably trapped by growth of olivine in the lake itself.

Conclusions

Inclusions in olivine from lunar and Hawaiian basalts can yield a variety of data on the temperatures of growth and the cooling history of the host crystals. The natural nucleation and growth of daughter crystals in melt inclusions provide relative cooling rates for different samples, and their melting in the laboratory within their olivine "bottles" provides data on the equilibrium temperatures, sequence, and nature of the phases other than olivine crystallized during cooling. Some constraints on the absolute natural cooling rates can be obtained from laboratory studies of nucleation in glassy inclusions. Not all types of data obtained are in agreement with other independent evidence, and several important questions remain unanswered. Regardless of this and the limitations imposed by the nature of the available inclusions, the implicit assumptions (particularly as to the representativeness of the sample), and the experimental techniques, some of these thermometric data are still valid, some cannot be obtained in any other way, and all must eventually be explicable.

Acknowledgements

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