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Gravitational Separation of Quenching Crystals: A Cause of Chemical Differentiation in Lunar Basalts

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Gravitational separation of quenching crystals: a cause of chemical differentiation in lunar basalts

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The low viscosity of lunar lavas allowed skeletal quenching crystals, which formed during post-eruption cooling of the flows, to sink and accumulate despite the low gravitational acceleration. The crystals which sank were not in chemical equilibrium with the liquid. Hand specimen bulk compositions are related to each other by differential movement of the materials composing the cores and rims of the skeletal 'phenocrysts', and not by any equilibrium chemical process. Crystal sinking must have occurred, and cumulates must have formed. Mafic hand specimens, consisting of skeletal phenocrysts in a more feldspathic groundmass, represent the cumulus enriched material and not the original liquids. The lava compositions were much more feldspathic, and were controlled by fractional crystallization at low pressures. The effects of such fractionation must be considered and, in most cases, restored before valid petrogenetic inferences about the mineralogy and chemistry of the lunar interior can be drawn.

Quenching crystals (crystals or parts of crystals which have formed during the cooling of a system, rather than those which equilibrated with the whole system at some constant temperature) are of common occurrence in experimental charges. In experimental charges such growths plus their groundmass are usually interpreted as evidence of the existence of an entirely liquid system of the appropriate bulk composition at the temperature of the experiment.

The inherent assumption in that interpretation is that the cooling rates are so fast that the skeletal crystals will not sink appreciably through the remaining liquid before its viscosity has reached some prohibitively high value due to the decline in temperature. That assumption has been extended (but has never been subject to the rigorous tests applied to lunar rocks which are described here) to terrestrial rocks, such as those described by Drever & Johnston (1957) or Viljoen & Viljoen (1969).

This paper examines whether the same assumption is justified for the lunar low titanium basalts of Oceanus Procelarum (Apollo 12) and Palus Putredenis (Apollo 15). Here the effects of the lower gravitational field on the Moon, which would make crystal sinking a less efficient process, might have been more than compensated by the much lower viscosity of lunar lavas (Weill *et al.* 1971).

The hand specimens of basalts from the Apollo 12 and 15 missions abound in skeletal or partially skeletal phenocrysts of olivine and pyroxene. Spinel phenocrysts were smaller and generally euhedral. The phenocrysts present are typically mantled, with cores of magnesian olivine, pigeonite and chromium spinel overlain by, respectively, more iron-rich olivine, by augite, and by titanium-rich spinel (for sources, see references in table 1). A petrographic examination of all the Apollo 12 hand specimens and many of the Apollo 15 specimens convinced us that the greater part of all material present in the cores, *as well as* in the rims of the phenocrysts, had crystallized during cooling of the system rather than in some period of effectively stable temperature. This interpretation would be supported by some petrographic accounts (Dowty *et al.* 1974) and by the results of controlled cooling rate experiments on this type of composition (Lofgren *et al.* 1974, 1975; Walker *et al.* 1975).

Many workers (Green *et al.* 1971; James & Wright 1972; Grove *et al.* 1973; Dowty *et al.* 1974) have advocated the further step of interpretation, reasonable in the light of experience with terrestrial rocks and experimental charges, that these skeletal textures indicate the original existence of liquids (lava flows) of the same composition as the hand specimens concerned.

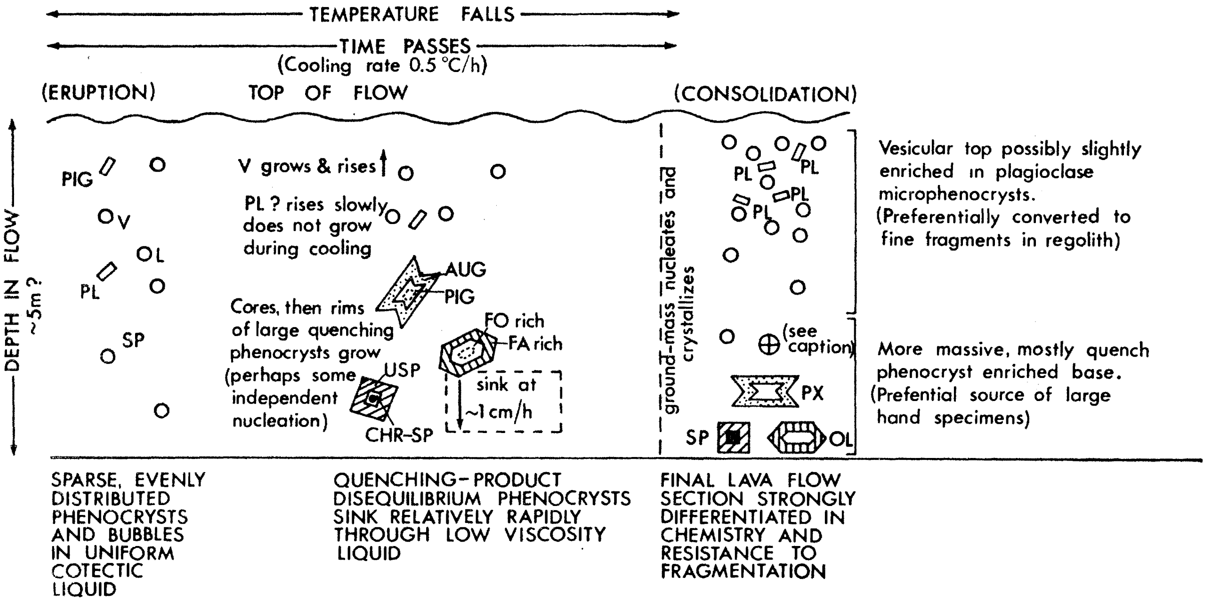


FIGURE 1. Model of the consolidation of a lunar lava flow (reproduced from O'Hara *et al.* 1975). Hand specimen compositions are related by the equation: $\text{rock}_1 = \text{rock}_2 \pm$ phenocryst cores ± phenocryst runs ± volatile components ± regolith contamination. ⊕ – explained in O'Hara *et al.* (1975).

The alternative hypothesis tested here (based upon reports by Biggar *et al.* 1972, 1974) is that the lavas erupted at both the Apollo 12 and 15 sites had compositions poorer in mafic mineral constituents and richer in potential plagioclase than those of the majority of the hand specimens. During cooling of the lavas, skeletal quenching crystals, comprising in varying proportions the cores and rims of the observed phenocrysts, formed and sank to the base of the original flows. The resultant rocks from the base of the flows would then contain abundant skeletal phenocrysts which might give rise to the misleading impression that the whole texture had developed from a liquid of the appropriate bulk composition. The hand specimens, it is suggested, come from large fragments exposed at the regolith surface, which were in turn derived preferentially from the more massive rocks richest in sunken phenocrysts and poorest in risen bubbles (see model, figure 1).

This hypothesis predicts or requires that:

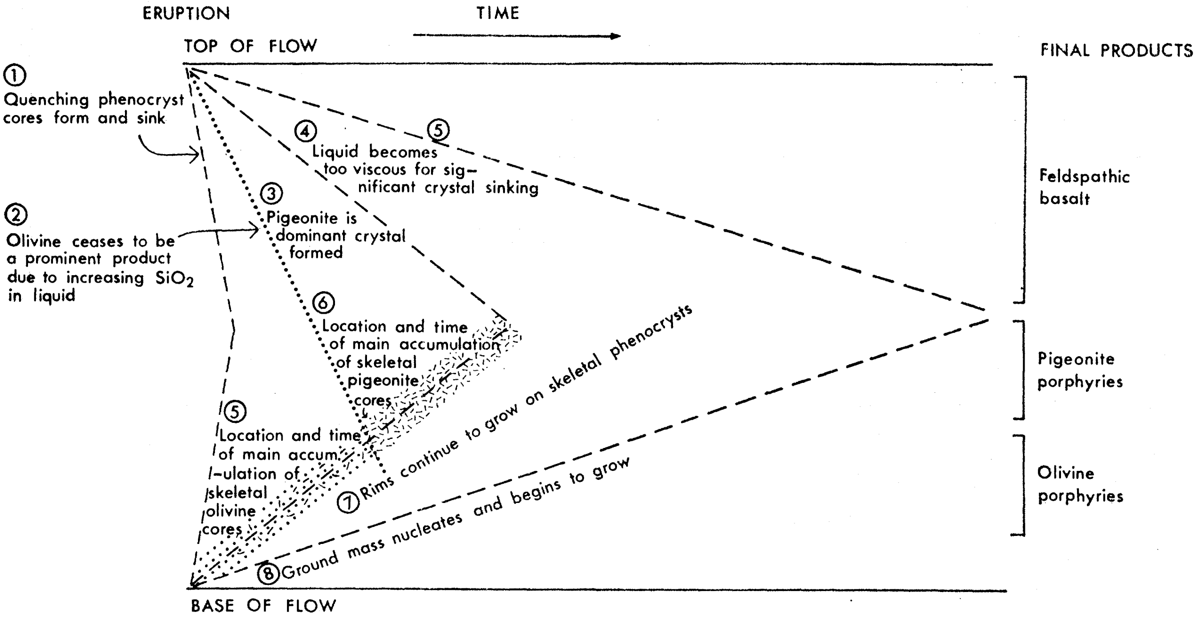
(1) The density and viscosity of the magma are such that crystals of the observed dimensions would have sunk rapidly relative to the increasing viscosity and the rate of advance of general crystallization fronts from the surfaces of the cooling flow.

(2) The chemistry of the individual hand specimens should be approximately related by:

$$\text{rock}_1 = \text{rock}_2 \pm \text{phenocryst cores} \pm \text{phenocryst rims}. \tag{1}$$

(3) When the composition inserted as 'rock$_2$' in equation (1) approximates to that of the original liquid, the proportions of additional phenocryst cores and rims required to produce each hand specimen composition (rock$_1$) should agree with the amount of phenocryst material observed in the petrography of rock$_1$.

(4) The average rock material available for fragmentation to form the regolith should be depleted in the components of olivine, pyroxenes and spinels relative to the average of the hand specimen compositions.



N.B. Cooling rates controlling crystallization of phenocryst rims, and the ground mass are widely variable, but controlled mainly by position within the flow. The cooling rate which controlled the phenocryst core formation was not necessarily controlled by the final position of that phenocryst.

FIGURE 2. Model of disposition of 'quenching' phenocrysts within a lunar lava flow as a function of time during consolidation, which permits pigeonite porphyries to form from the same initial magma as the olivine basalts, due to the early failure of olivine to continue forming from the increasingly siliceous residual liquids. Formation of the pigeonite and olivine porphyries from the same flow is *not*, however, an essential feature of the thesis presented here.

(5) Regolith thickness should have been less than, or of the same order of size as the flow thicknesses. Larger hand specimens would then have been preferentially derived from near the base of the top flow. The bias towards more mafic compositions among larger hand specimens might, however, have been assisted by the greater vesicularity, or lower resistance to fragmentation of the less porphyritic rocks (see figure 1).

Each of these propositions is examined in more detail below.

(1) Walker *et al.* (1975) have demonstrated experimentally that suitable skeletal mantled phenocrysts would have formed in low titanium basalt magma by a temperature of 1075 °C, at a cooling rate of *ca.* 0.5 °C/h. At that temperature the viscosity of the system is still sufficiently low that crystals of the size observed in the natural rocks would have sunk through several metres before the flows consolidated (Walker *et al.* 1975).

Unless cooling of the flows occurred exclusively through the lower surfaces, a most improbable model, it follows that sinking of the skeletal phenocrysts and, therefore, the accumulation of the phenocrysts at some point, *must have occurred.* A model of what occurred is advanced in figure 2. If the original magma composition lay close to the limit of olivine saturation under the cooling conditions, the lowest cumulus-enriched horizons may be olivine-porphyries, but pigeonite-porphyries will predominate at a higher level. (This is in accord with the petrography of specimens from the Apollo 12 and 15 sites.)

More than one lava flow was penetrated during the formation of the regolith at each of the sites. There is, therefore, no adequate model which excludes from the sample some of the rocks into which quenching phenocrysts *must have* accumulated. If the most mafic hand specimens such as 12002, 12040 (i.e. those richest in the quenching phenocrysts) are chosen as representative of the liquid composition which was erupted, there remain no samples which can be identified with the cumulates. If, however, the magma composition is equated with the average rock material converted to regolith (see 3, 4 below), the mafic hand-specimen samples fulfil the anticipated petrographic and chemical role of the cumulus-enriched samples.

TABLE 1. SOURCES OF PHASE DATA USED IN FOLLOWING CALCULATIONS

group I	group II	group III
sources of mineral analyses		
Olivine core 12020 (1, tbl 2, no 1)	12065 (3, tbl 3, trav 11/1, no 1)	12022 (5, tbl 1, no 7)
Olivine rim 12020 (1, tbl 2, no 2)	12065 (3, tbl 3, trav 11/1, no 2)	12022 (5, tbl 1, no 8)
Pyroxene core 12020 (1, tbl 1, no 1)	12065 (3, tbl 1, trav 9/1, no 6)	12063 (3, tbl 2, trav 2/1, no 1)
Pyroxene rim 12020 (1, tbl 1, no 2)	12065 (3, tbl 1, trav 9/1, no 1)	12022 (5, tbl 1, no 15)
Spinel core 12020 (2, tbl 2)	12065 (4, tbl 2, no 5A)	12022 (4, tbl 2, no 3)
Spinel rim 12020 (2, tbl 1)	12065 (4, tbl 2, no 5)	12022 (4, tbl 3, no 1)

1, Kushiro *et al.* (1971); 2, Haggerty & Meyer (1970); 3, Hollister *et al.* (1971); 4, Cameron (1971); 5, Weill *et al.* (1971).

Rock analyses in tables 2–4 identified by letter after sample number, as in James & Wright (1972, table 5); C, Compston *et al.* (1971); E, Engel *et al.* (1971); H, Kushiro & Haramura (1971); 12065 H1 is rock, 12065 H2 is exterior chip; M, Maxwell & Wiik (1971); WL, Willis *et al.* (1971).

(2) The caption to figure 1 explains the chemical relationships which should be observed if sinking of quenching phenocrysts has occurred. The approximate form of the relationship given in equation (1) above yields the predicted excellent description of the chemistries of the different specimens (tables 2–4) with residuals below the limits of precision of the analyses for all the non-volatile elements. Earlier Biggar *et al.* (1971) suggested that three different flows might be present at the Apollo 12 site. The basis for a separation of a pigeonite-porphyry flow from an olivine-porphyry flow no longer exists in the light of these new considerations, but the more titaniferous basalts treated in table 4 probably come from a different flow.

(3) The precise amounts of the core and rim phenocryst materials which are present in each of the samples are not known. The amounts of the different phenocryst materials which must be added to the least porphyritic rocks in order to produce the more mafic hand specimen compositions (tables 2–4) are, however, consistent with the amounts of phenocryst materials recorded in the available petrographic descriptions.

The high proportion of the rim materials relative to the core materials which are required by the calculations are consistent with the observed petrography. (An apparently thin rim around the outside of a crystal contributes a high proportion of its total volume.) The rims described or figured in the sources quoted in table 1 would support rim:core ratios as high as 7:1. This latter feature is most important when considering the consequences of the quenching crystal differentiation process.

Table 2. Best fits to equation (1), using a modified Wright-Doherty (1970) program

(Rock analysis, in left hand column = the percentages of another stated rock analysis (column 2) plus the amounts of olivine, pyroxene and spinel phenocrysts given in the next six columns. Residuals for each oxide are listed in the next 11 columns, and the sum of squares of residuals in the final column. Values in italics are negative. The mineral data came from group I (olivine porphyries) of table 1, but both olivine and pigeonite porphyries have been fitted to these data. 12038 is a rock very close in composition to the plagioclase saturated cotectic liquid; misfits are largely connected with the Na_2O content. Sodium is very volatile at the eruption temperature of these rocks.)

analysis of rock sample =		olivine		pyroxene		spinel		residuals											
	rock	core	rim	core	rim	core	rim	SiO_2	TiO_2	Al_2O_3	Cr_2O_3	FeO	MgO	CaO	MnO	Na_2O	K_2O	P_2O_5	ΣR^2
Fits to 12038 H																			
(*a*) Olivine porphyries																			
12002 WL	60.36	22.49	3.99	0.63	10.05	0.97	1.51	.00	.00	.01	*.01*	.00	.00	*.01*	.01	*.16*	.01	.10	.038
12004 C	60.88	12.88	5.74	3.09	15.29	*0.14*	2.26	.00	.00	.01	.00	.00	.00	.00	.01	*.19*	.03	.07	.047
12004 M	66.30	11.48	3.04	9.80	7.46	0.32	1.60	.01	.01	.02	.00	.01	.01	.00	.01	*.19*	.02	.04	.045
12009 C	66.50	9.10	4.57	5.21	12.85	*0.05*	1.83	.01	.01	.02	.00	.01	.01	.00	.01	*.19*	.01	.06	.046
12018 H	65.04	25.61	1.68	1.10	5.05	0.38	1.14	.04	.03	.04	.02	.03	.03	.02	.00	*.22*	.00	.01	.054
12020 H	61.73	21.14	2.57	2.84	10.21	0.36	1.14	.03	.02	.04	.02	.02	.02	.02	.00	*.19*	.02	.01	.039
12035 C	64.03	28.33	5.65	5.89	7.31	0.21	0.36	.01	.01	.02	.00	.01	.01	.01	.03	*.20*	.01	.05	.047
12040 H	56.43	27.49	2.15	1.54	10.53	0.33	1.53	.03	.02	.04	.02	.03	.03	.02	.00	*.20*	.01	.01	.046
12075 E	74.15	17.61	1.43	8.40	*2.21*	*1.05*	1.67	.01	.00	.02	.00	.00	.00	.00	.00	*.17*	.00	.15	.051
12075 R	60.87	17.97	2.91	7.07	9.70	0.18	1.30	.01	.01	.02	.00	.01	.01	.00	.00	*.16*	.03	.07	.031
(*b*) Pigeonite porphyries																			
12064 H	85.42	*4.04*	3.32	*5.69*	18.89	*0.76*	2.87	.05	.03	.06	.03	.04	.03	.03	.00	*.28*	.01	.00	.087
12021 H	82.42	*2.21*	3.31	1.11	16.16	*0.45*	1.88	.03	.02	.04	.02	.03	.02	.02	.00	*.18*	.01	*.01*	.036
12052 H	80.55	*0.25*	3.53	3.54	11.35	0.04	1.22	.04	.03	.05	.02	.03	.03	.02	.00	*.24*	.01	.01	.062
12065 H1	85.24	0.69	3.19	0.12	9.47	*0.15*	1.45	.05	.04	.06	.03	.04	.04	.03	.00	*.30*	.01	.00	.099
12065 H2	83.37	*0.72*	2.96	0.78	12.70	*0.01*	0.92	.00	.00	.01	*.01*	.00	.00	*.01*	.00	*.19*	.02	.19	.075
(*c*) Olivine porphyry to olivine porphyry																			
Fits to 12018 H																			
12040 H	86.77	5.28	0.70	0.57	6.15	*0.01*	0.55	.00	.00	.00	.00	.00	.00	.00	.00	*.01*	.01	.00	.000
12020 H	94.89	*3.16*	0.99	1.79	5.44	0.00	0.06	*.01*	.00	*.01*	.00	*.01*	.00	.00	.00	.02	.01	.00	.001
Fits to 12020 H																			
12040 H	91.43	8.18	0.20	1.11	1.21	*0.01*	0.49	.01	.01	.01	.00	.00	.00	.00	.00	*.03*	.00	.00	.001

TABLE 3. DATA AND LAYOUT AS FOR TABLE 2. MINERAL DATA CAME FROM GROUP II (PIGEONITE PORPHYRIES) OF TABLE 1

(12064 is the pigeonite porphyry with the fewest phenocrysts in this group. Values greater than 100% arise in the second column when rock$_1$ is closer to the liquid composition than rock$_2$.)

analysis of		olivine		pyroxene		spinel		residuals											
rock sample =	rock	core	rim	core	rim	core	rim	SiO_2	TiO_2	Al_2O_3	Cr_2O_3	FeO	MgO	CaO	MnO	Na_2O	K_2O	P_2O_5	ΣR^2
Fits to 12064 H																			
(*a*) Olivine porphyries																			
12018 H	77.68	39.62	*11.00*	0.78	*6.77*	0.64	*0.94*	.00	.00	.00	.00	.00	.00	.00	*.01*	.02	.00	.00	0.000
12018 R	77.28	37.66	*9.04*	*1.95*	*3.76*	0.73	*0.92*	*.02*	*.02*	*.02*	*.02*	*.02*	*.02*	.02	.02	.07	*.01*	.05	0.011
12018 C	66.44	20.20	5.92	6.38	0.74	0.27	0.05	*.02*	*.02*	*.02*	*.02*	*.02*	*.02*	*.02*	.00	.01	.01	.06	0.008
12020 C	62.50	9.28	11.14	10.09	6.27	0.26	0.46	*.03*	*.02*	*.02*	*.02*	*.02*	*.02*	*.02*	.00	.02	.02	.07	0.012
12020 H	72.90	29.09	*4.17*	4.24	*1.85*	0.63	*0.83*	.00	.00	.00	.00	.00	.00	.00	*.01*	.02	.01	.01	0.001
12020 R	73.56	24.67	*1.60*	6.08	*2.45*	0.49	*0.76*	*.02*	*.01*	*.02*	*.01*	*.02*	*.02*	*.01*	.00	.04	.01	.07	0.007
12040 C	63.80	28.79	0.55	4.93	2.00	0.31	*0.38*	*.02*	*.01*	*.01*	*.01*	*.01*	*.02*	*.01*	.01	.02	.00	.05	0.006
12040 H	66.66	38.29	*7.52*	3.31	*1.02*	0.61	*0.33*	.00	.00	.00	.00	.00	.00	.00	.00	.00	.00	.01	0.000
12040 S	67.65	36.43	*9.66*	6.97	*2.46*	0.59	0.49	*.02*	*.02*	*.02*	*.02*	*.02*	*.02*	*.02*	.00	.04	*.01*	.06	0.011
(*b*) Pigeonite porphyries																			
12021 H	96.62	0.85	1.18	2.91	*0.91*	0.13	*0.79*	*.01*	*.01*	*.01*	*.01*	*.01*	*.01*	*.01*	.00	.09	.00	*.01*	0.009
12065 H1	100.96	5.44	*0.35*	1.39	*6.54*	0.34	*1.25*	.00	.00	.00	.00	.00	.00	.00	.00	*.02*	.00	.00	0.000
12065 H2	98.28	4.02	*0.61*	3.15	*3.58*	0.41	*1.68*	*.05*	*.03*	*.04*	*.03*	*.04*	*.04*	*.04*	.00	.08	.01	.19	0.054
12052 H	94.93	1.12	3.38	5.10	*3.70*	0.48	*1.30*	*.01*	*.01*	*.01*	.00	*.01*	.00	*.01*	.00	.02	.00	.01	0.001
(*c*) Pigeonite porphyry to pigeonite porphyry																			
Fits to 12021 H																			
12065 H1	104.47	4.54	*1.59*	*1.63*	*5.57*	0.21	*0.42*	.02	.01	.02	.01	.02	.01	.01	.00	*.11*	.00	.01	0.014
12065 H2	101.73	3.15	*1.81*	0.18	*2.65*	0.28	*0.88*	*.04*	*.02*	*.03*	*.02*	*.03*	*.03*	*.03*	.00	*.01*	.01	.20	0.046
12052 H	98.26	0.28	2.22	2.22	*2.80*	0.35	*0.53*	.01	.00	.01	.00	.01	.00	.00	.01	*.07*	.00	.02	0.005
Fits to 12065 H1																			
12052 H	94.04	4.00	3.72	3.78	2.45	0.15	0.13	.01	.01	.01	.01	.01	.01	.01	.00	.04	.00	.01	0.002

Table 4. Data and layout as for table 2. Mineral data came from group III (the more titaniferous basalts) of table 1

(Only the more titaniferous basalts have been fitted. 12051 is the least porphyritic specimen of the group, and is closest to the plagioclase saturated cotectic liquid compositions.)

analysis of rock sample =	rock	olivine		pyroxene		spinel		residuals											
		core	rim	core	rim	core	rim	SiO_2	TiO_2	Al_2O_3	Cr_2O_3	FeO	MgO	CaO	MnO	Na_2O	K_2O	P_2O_5	ΣR^2
Fits to 12051 M																			
12022 H	91.90	7.86	15.38	*10.54*	*5.68*	*0.34*	1.43	.02	.01	.02	.01	.02	.02	.02	*.02*	.01	.03	*.04*	.014
12022 E	92.10	5.55	8.83	*1.67*	*6.53*	*4.81*	6.53	*.03*	*.02*	*.03*	*.02*	*.02*	*.02*	*.02*	*.02*	.19	.03	.07	.053
12063 WL	92.49	14.57	*2.23*	*9.49*	2.78	*2.15*	4.04	*.02*	*.01*	*.02*	*.01*	*.02*	*.02*	*.02*	.00	.02	.02	.07	.008
Fits to 12051 C																			
12022 H	91.03	3.14	16.55	*4.34*	*7.97*	*0.60*	2.18	.02	.02	.02	.02	.02	.02	.02	*.02*	.04	.00	*.06*	.015
12022 E	91.24	0.81	10.01	4.56	*8.83*	*5.07*	7.28	*.03*	*.02*	*.03*	*.02*	*.02*	*.02*	*.02*	*.02*	.22	.00	.05	.059
12063 WL	91.64	9.78	*1.01*	*3.23*	0.45	*2.41*	4.79	*.02*	*.01*	*.01*	*.01*	*.01*	*.01*	*.01*	.00	.05	*.01*	.06	.007
Fits to 12063 WL																			
12022 H	99.32	*6.56*	17.53	*1.12*	*8.40*	1.79	*2.56*	.04	.03	.04	.03	.04	.03	.03	*.02*	*.01*	.01	*.12*	.031
12022 E	99.57	*8.93*	11.02	7.77	*9.27*	*2.67*	2.52	*.01*	*.01*	*.01*	.01	*.01*	*.01*	*.01*	*.03*	.16	.01	*.01*	.036

(4) The average mare-derived impact-melted basalt glass fractions in soils from most of the investigated sites (low and high titanium basalts) are more feldspathic than (i.e. depleted in constituents of olivine, pyroxene and spinel relative to) the majority of the hand specimens (Reid & Jakes 1974). These average mare derived basalt glass compositions closely resemble the average target rock compositions which were converted to regolith at the Apollo 11 and 12 sites at least (Biggar *et al.* 1972; O'Hara *et al.* 1974). The proportion of highland derived materials in the soils is too low to account for the more feldspathic character of the regolith in terms of simple mixture with picritic hand specimen materials. The average contribution expected from highland sources (taken to be represented by highland soil compositions) is moreover too poor in silica to provide suitable chemical mixtures (figure 3), even if it could be argued that a larger highland contribution were occult in impact-melted mixtures of picritic and highland-derived materials. Lithic fragments, identified by their petrography as being pieces of crystalline basalts, have average compositions which resemble the average target rock contributing to the regolith, proving the existence of a rock of this composition which crystallized with the characteristic thermal history of the basalts rather than that of impact melts.

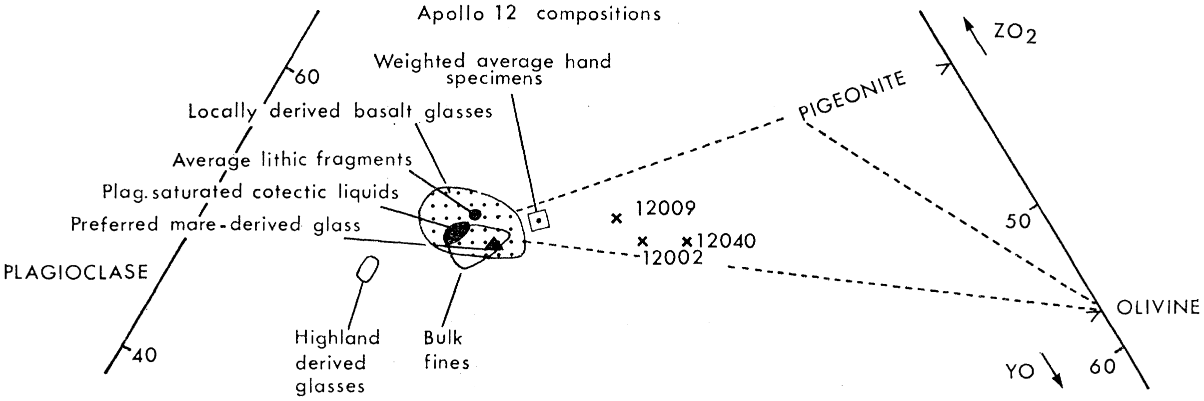


FIGURE 3. Relationships between hand specimen, soil and highland contaminant compositions at the Apollo 12 site to show the unlikelihood of samples such as 12002, 12009, 12040 representing the parental magma composition (modified from Biggar *et al.* 1972, Figure 16). The projection is from 'diopside' into the plane anorthite-olivine-silica of the representation tetrahedron XO-YO-R_2O_3-ZO_2 (Jamieson 1970).

Exceptions to this generalization are some soils from the Apollo 15 site which fall along a mixing line between picritic hand specimens and highland-derived components. However, there is abundant evidence of extensive magma drainage from this site (Howard *et al.* 1972) and the relationships would be fully explained if skeletal quenching crystals had accumulated into the base of the uppermost flow at least, before the removal of the overlying liquid (close to the average magma in composition) by drainage. Reid & Jakes (1974) again found a feldspathic mare basalt as the main contributor of small glass particles at this site, presumably reflecting sampling from a wider area. As shown by the calculations and by data projections (figure 4), the Apollo 15 hand specimens are chemically related to more feldspathic liquids by addition of the skeletal phenocryst materials which they actually contain. The two groups cannot be related *directly* to each other by any crystal differentiation process involving the phenocrysts present but are readily interrelated via the plagioclase-saturated cotectic liquid. The alternative interpretation, that both picritic and pyroxene phyric basalts at this site underwent just sufficient *in situ* quenching growth of skeletal phenocrysts, to create this confusing impression, is less than satisfactory, because such crystals would have sunk and accumulated somewhere (see 1 above).

The most reasonable interpretation of all the facts is that the hand specimen compositions are richer in components of olivine, pyroxene and spinel than was the average rock composition converted to regolith over most of the maria surfaces. The main contribution to that average rock composition were the crystallization products of a basaltic liquid which had yielded the picritic samples by local accumulation of skeletal phenocrysts.

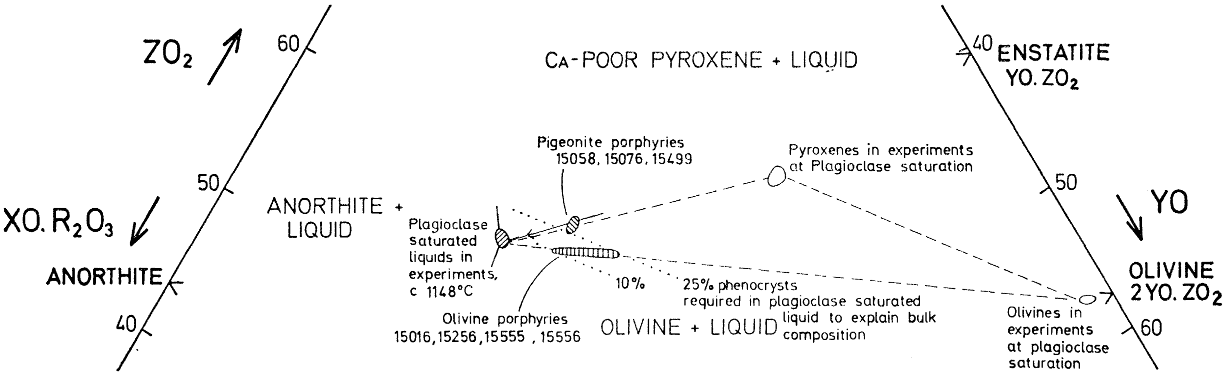


FIGURE 4. Compositions of Apollo 15 olivine and pigeonite porphyries in relation to the plagioclase-saturated cotectic liquid from which it is suggested, both have formed by sinking of 'quenching' phenocrysts. Projection from 'spinel' (the other crystal species present in the experimental crystallization) into the plane of anorthite-olivine-silica of the representation tetrahedron XO-YO-R_2O_3-ZO_2 (Jamieson 1970). N.B. This projection, based on experimental data by Humphries *et al.* (1972 and unpublished) does not distinguish between core and rim material in the phenocrysts – all olivines project at one point.

(5) Regolith thicknesses in the maria at the Apollo 12 and 15 sites were ~5 m (Howard *et al.* 1972). The thicknesses of individual lava flows on the lunar surface, estimated from heights of flow fronts and from direct observation in the walls of Hadley Rille were 10–60 m (Howard *et al.* 1972; Schaber 1973).

Regolith thicknesses may, therefore, have been comparable to or substantially less than the flow thicknesses, in which case the material originally forming the upper part of the flow unit will have undergone a greater amount of 'gardening' by meteorite impact, and will, therefore, have contributed preferentially to the smaller fragments. While this might appear superficially to favour a model in which the cumulate-enriched portions of the flows were not sampled during regolith formation, such a model is unacceptable because the required highly mafic fragments have not been recovered in any proportion, even from boulders excavated from depths of over 90 m (Swann *et al.* 1972; Warner 1971) by large craters.

Many of the hand specimens are vesicular, and it would be logical to expect that any bubbles present would have risen rapidly through the flow unit while denser crystals were sinking. Even more vesicular upper sections to the original flow units must, therefore, have been formed as surely as the massive, cumulus-enriched portions already discussed. An increase in the proportion of vesicles would increase the probability that the material would become fragmented into smaller particles during repeated meteorite bombardment.

The five tests of the hypothesis discussed above are either neutral or affirmative; the alternative explanations of the facts discussed in tests 2 to 5 are less plausible than the hypothesis advocated. There are, however, two observations which might still be held to favour one of the alternative interpretations:

Mg/Fe ratio of early olivine crystals

It has been argued that the Mg/Fe ratios of the crystals developed in experiments at the liquidus of hand specimen samples 12065, 12021 (Green *et al.* 1971) predicted to be present in 12002 (Grove *et al.* 1973) or observed to be present in 12002 (Walker *et al.* 1975) are the same as those of the most magnesian olivines present in the natural rocks; the olivines must, therefore, have begun their crystallization when the samples were all liquid.

Biggar *et al.* (1974) suggested that during rapid growth of olivines, the equilibrium distribution coefficient of iron and magnesian between olivine and liquid might not be maintained. This possibility has been disproved in experiments by Walker *et al.* (1975). The argument presented above is, therefore, valid *provided that* the experimental observations were valid, i.e. that they refer to appropriate bulk compositions, and make fair comparisons between like observations.

The experiments by Green *et al.* (1971), and Walker *et al.* (1975) were conducted in iron capsules. If the material loaded into the capsules was not in bulk equilibrium with iron metal at the liquidus temperature, excess oxygen in the charge would react with the container, producing additional FeO to dissolve in the melt. This effect is probable in the case of the experiments on 12065 (see O'Hara *et al.* 1975) because of the routine procedure for charge preparation adopted by Green *et al.* (1971). The experiments of Walker *et al.* (1975) were carried out in pure iron capsules in sealed silica tubes, where it is probable that oxygen diffused slowly through the wall of the silica tube (see Gray *et al.* 1974, p. C106) and caused oxidation of the container into the charge (reaction will occur at the charge-container interface at a lower oxygen fugacity (O'Hara *et al.* 1975) than is required to oxidize the pure iron capsule itself).

In both cases the olivine crystals formed at the liquidus will have equilibrated in a system of lower Mg/Mg + Fe than is appropriate. The resultant olivine compositions may be as iron rich as those appropriate to a much lower temperature in the true natural rock system. O'Hara *et al.* (1975, figure 4) present further arguments suggesting the operation of this process.

Similar arguments would still apply to the situation at lower temperatures – the olivines generated in the experiments would always be more iron-rich than those appropriate to the natural crystallization at the same temperature. Similar arguments again may apply to the experiments by Biggar *et al.* (1971, 1972) in which all the iron present originally as metal and as sulphide probably appears as FeO in the final run products.

It is not suggested that these small increments in the FeO content of the charges seriously alter the broad outlines of the phase equilibria reported for the samples – the orders of appearance of the major phases are the same in all reports, as are the temperatures of appearance of the various minerals (within stated limits of control and precision). They may, however, be large enough, and are certainly in the right direction, to invalidate detailed comparisons which turn finally upon differences of no more than 7 mol % of forsterite molecule in the olivine (see O'Hara *et al.* 1975, figure 4).

It is extremely difficult to conduct experiments upon the low titanium basalts under conditions which reproduce *exactly* the iron oxide content of the silicate portion, and the other conditions of crystallization of the natural rock. The inclusions of silicate liquid which were trapped and completely enclosed within the olivine crystals during their growth may afford a more readily controlled closed system in which to follow the relation between olivine composition and temperature. Roedder & Weiblen (1971) reheated the silicate liquid inclusions in natural olivines from rock 12018. They found that on heating to 1220 °C, over 100 °C below the liquidus temperature of the hand specimen composition, the olivine adjacent to the silicate melt had *already* become more magnesian than the most magnesian olivine reported from the natural rock. The implication, directly contradicting the conclusions from experimentation in iron capsules, is that the olivine phenocrysts in 12018 began crystallizing at a temperature significantly below 1220 °C, when the liquid composition would have been close to that of the plagioclase-saturated liquids which match the average rock composition contributing to the regolith (Biggar *et al.* 1971, 1972).

Finally, if the argument contained in this paper is accepted, the present bulk compositions of samples such as 12065, 12002 have been subject to additions of iron-rich olivine and pyroxene, in the form of phenocryst rim materials, during a disequilibrium chemical process. This process automatically vitiates any argument based upon observation of compositions of crystals present at equilibrium at the liquidus.

In our opinion the argument based upon comparison of natural olivine compositions with those found in experimental charges is not established because the bulk compositions of the charges are not truly relevant to the situation which obtained in the natural rocks, both by virtue of problems of experimental control and by virtue of the sinking of mantled, disequilibrium phenocrysts. The more reliable evidence, from a silicate liquid inclusion sealed in its own olivine container favours the hypothesis advocated in this paper.

Rb/Sr ratio and initial $^{87}Sr/^{86}Sr$

Individual hand specimens of Apollo 12 lavas yielded different Rb/Sr ratios, and had different $(^{87}Sr/^{86}Sr)_I$ ratios (Compston *et al.* 1971).

If these results are interpreted on the simplest possible closed system model they indicate that at least six different lavas were sampled, each derived from a different region of the lunar mantle with its own characteristic Rb/Sr and $^{87}Sr/^{86}Sr$ ratios at the time of partial melting. Such results apparently support the James & Wright (1972) interpretation of the major element chemistry, the observed differences of the ratios being impossible to produce by differential movements of olivine, pyroxene or spinel. The chemical and petrographic fits discussed in test 2 above must then be regarded as a remarkable coincidence.

However, two considerations indicate the need for caution. Few similarly detailed studies have been published of samples from different parts of what is known to be a single lava flow on the Earth. Such studies are necessary as a control to establish that differences similar to those seen in the Apollo 12 samples do not arise from some unsuspected mechanism, and are not commonplace in single basalt flows. In one terrestrial example which has been studied in detail (Brookins *et al.* 1975), $(^{87}Sr/^{86}Sr)_I$ varies from 0.7037 to 0.7081, and Rb/Sr from 0.051 to 0.070, a range of values as great as or greater than that used by Compston *et al.* (1971) to subdivide the different 'flows' of the Apollo 12 site.

In the second place the Apollo 12 lavas flowed out over a surface which, if it was covered by a regolith comparable in composition with that now present on top of the final flows, contained eight times more Rb and twice as much Sr than the lava itself due to the presence of a small amount of ancient 'KREEP' rich in those elements. The high Rb/Sr ratio in the KREEP component ensured that the Sr of the regolith had a high $^{87}Sr/^{86}Sr$ ratio at the time of eruption of the Apollo 12 lavas. Variable contamination of different parts of the lava flows by incorporation of from 5 to 15 % of the regolith over which they flowed would be sufficient to produce the observed range of Rb/Sr and $(^{87}Sr/^{86}Sr)_I$ but would produce no readily observed change in the major element composition. Even smaller amounts of selective contamination by KREEP-rich material would produce similar effects.

12009 *again*

12009 is unquestionably the most favourable hand specimen sample on which to base an argument for the existence of picritic liquids at the lunar surface. It differs from the other samples under discussion in three main respects:

(i) it was cooled much more rapidly and apparently from a much higher temperature than the other samples, such that the only skeletal phenocrysts formed were of olivine, and the groundmass remained as glass.

(ii) the glass composition now host to the skeletal phenocrysts cannot represent an equilibrium liquid composition, and does not closely resemble the average magma composition (Biggar *et al.* 1972, figure 5).

(iii) the major element chemistry of 12009 relates its composition to that of the other rocks at the site by relative movement of the mantled phenocrysts (table 2) but in amounts which do not correspond with the observed petrography.

Impact melting *must* have occurred when the other picritic hand specimens were excavated. The process is known to have produced small glass beads of picritic glass in the soils. More extensive coatings or globs of impact generated melts are known to have formed, on 12017 for example. The distinctive thermal history of 12009 (rapid quenching from a very high temperature) is common to all the impact melted glasses, and the development of glasses metastably depleted in olivine constituents as observed in 12009 (Biggar *et al.* 1972) during rapid quenching growth of olivine is a familiar feature of experimental charges also (Cawthorn *et al.* 1974).

Accordingly, while 12009 must be accepted as a quenched picritic liquid, it is not a magma of internal origin, but an impact generated melt. It has no direct relevance to the discussion of either the petrogenesis of the lavas, or the nature and processes of the lunar interior.

Consequences of the chemical differentiation by sinking of quenching crystals, and resultant confusion about the liquid composition

The hand specimens do not represent the compositions of liquids emanating from the lunar interior, nor those of crystal-liquid mixtures determined by processes occurring within the Moon. The hand specimen compositions are not even related by equilibrium chemical processes to the real liquid compositions which were erupted. The greatest caution must be exercised, therefore, when developing petrogenetic arguments based upon the details of the chemistry of these rocks.

For example, figures 5 and 6 show one effect of the process, and its consequences for petrogenetic interpretation.

Original magma (liquid) B develops during cooling phenocrysts whose cores are of composition O_c, but which have much more abundant rim material O_R mantling them. Sinking of these crystals leads to accumulates such as A forming, accompanied by residual magma. (During quenching differentiation, the trend of liquid evolution does not follow any path predictable from the equilibrium phase relationships; see Walker *et al.* (1975).) Rock A cannot be produced from mixtures of B or the residual magna with the original (potentially equilibrium) cores of the olivine crystals O_c. If the process operating were not appreciated, this might lead to the erroneous conclusion that rock A and the residual liquid are unrelated. Such a 'conclusion' might appear to hold for all pressures and all forms of the equilibrium phase relationships.

The equilibrium experimental crystallization of sample A would yield liquidus olivine O_M, and subsequent olivine compositions between O_M and O_E, while the residual liquids develop along the curved path A–B_E, at which temperature pyroxene would begin to crystallize (below the temperature at which pyroxene was present in liquid B). At no point in the experimentally realizable trajectory does the state of the whole system, or that of its liquid part, reproduce a condition relevant to the real magma B.

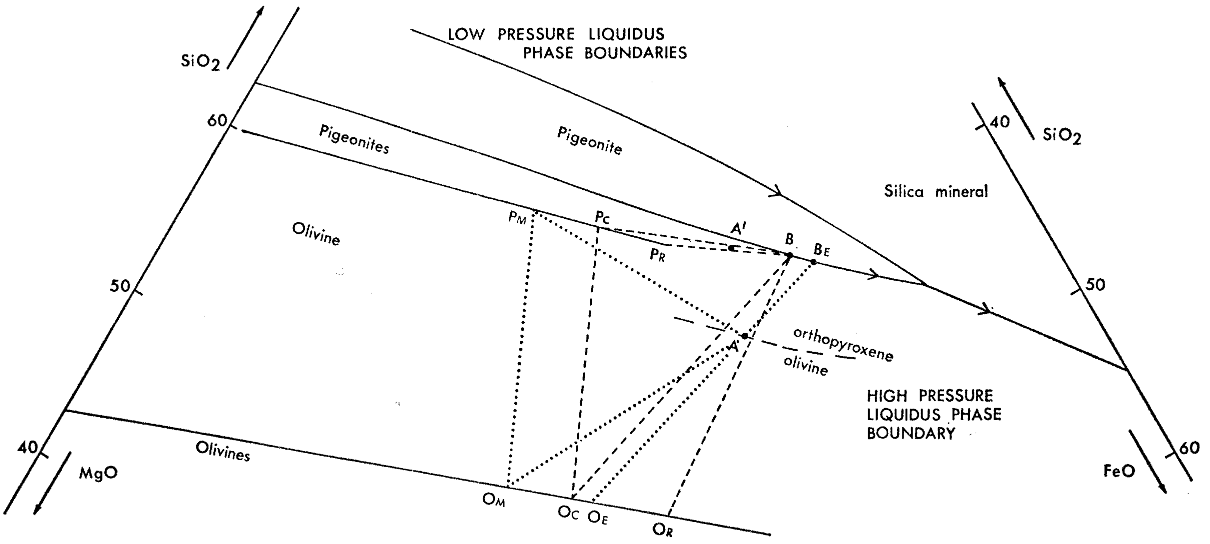


Figure 5. Petrogenetic effects of the accumulation of quenching phenocrysts, illustrated in terms of variations in MgO, FeO, and SiO_2.

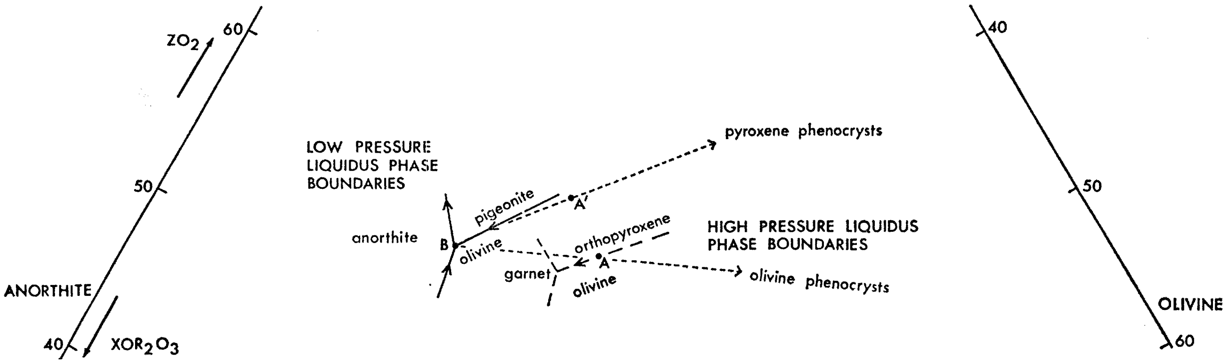


Figure 6. Petrogenetic effects of the accumulation of quenching phenocrysts, illustrated in terms of variations in the system plagioclase-olivine-silica.

It follows that results of phase equilibria studies carried out on specimen A have only a broad general relevance to the crystallization processes in the real magma B. No weight should be attached to detailed comparisons between natural and experimentally produced olivines (it is possible for O_M to be more or less magnesian than O_c).

The situation is made yet more complex if the role of differential accumulation (in accordance with figure 2) of pyroxene cores and rims is considered, or if the effects of loss or gain of iron oxide due to unavoidable reactions between silicate melt and container are taken into account (see for example Biggar *et al.* 1974, figure 3).

Composition A was not a liquid; but the consequences of treating it as a liquid (on the strength of the presence of skeletal phenocrysts which are assumed, erroneously, to have formed *in situ*) and of assuming that it had erupted unchanged from some deep region within the Moon, are illustrated in figures 5 and 6.

There exists some high pressure and a given temperature (which may be correlated with a given depth within the Moon) at which composition A lies exactly at the interface (broken line) between the liquidus fields of olivine and pyroxene. In the simplest possible model, a liquid of composition A might be a partial melt derived from an olivine–pyroxenite source rock at that depth, the temperature of partial melting being given by the liquidus temperature of A and the compositions of residual olivine and pyroxene in the lunar mantle (O_M, P_M) by the nature of the liquidus crystals in A at that pressure.

By further assumptions about the ratio of olivine to pyroxene in the residuum (which cannot be defined by experiments on A) and about the proportion of partial melt formed, a 'unique' composition of the lunar mantle may be assigned within the region O_m–P_m–A.

All this, it will be appreciated, is an academic exercise. A was not a liquid, and the composition which is relevant to any such exercise is the liquid B.

Figure 6 illustrates a further aspect of the problem. Choice of A as the relevant liquid instead of composition B means that there is no pressure at which liquid A could be in equilibrium with crystals of feldspar (whereas B as drawn is in simultaneous equilibrium with feldspar, olivine and pyroxene at low pressure).

If liquid B has developed a marked relative deficiency in some trace element, such as europium, due to the prior fractionation of feldspar from the system, that deficiency will still be apparent in composition A (because the addition of olivine, pyroxene, or spinel to a sample of liquid B will have a negligible effect on the concentration of europium relative to the other rare earth elements). However, whereas the europium deficiency is readily explicable if the magma had composition B, it cannot be explained by any equilibrium process affecting composition A. Adoption of A as the magma composition immediately creates a need for disequilibrium processes within the lunar mantle, or models in which late stage mafic cumulates at depth within the Moon become remelted (in preference to more plagioclase-rich cumulates which would have had lower temperatures of melting!).

Petrogenesis of mare-surface lavas

The average rock composition present at the Apollo 12 site is present as a non-porphyritic crystalline basalt. Liquids of this composition are in simultaneous equilibrium with olivine, calcium-poor pyroxenes, chrome-spinel and plagioclase at their liquidus at low pressure, i.e. they are low pressure plagioclase-saturated cotectic liquids (Biggar *et al.* 1971, 1972).

Similar low-titanium basalt compositions are widespread on the maria (Reid & Jakes 1974), and a similar liquid is a suitable parent (prior to magma drainage) for the hand specimens collected from the Apollo 15 site (Humphries *et al.* 1972).

The average compositions of high titanium basalts collected from maria surfaces at the Apollo 11 and 17 sites are also those of low pressure plagioclase-saturated cotectic liquids, but in simultaneous equilibrium with ilmenite, augite and in some cases armalcolite, in addition to plagioclase, olivine, calcium poor pyroxene and a chrome-titanium spinel (O'Hara *et al.* 1974, 1975; O'Hara & Humphries 1975).

Low pressure multiphase cotectic liquid characteristics can be imposed upon magmas by any process which leads to effective crystal-liquid fractionation at low pressure. (It is the first duty of experimental petrology to test for such influences.) The topographic environment of the maria suggests the existence of large lava lakes within which slow cooling and fractional crystallization would have produced, as a mandatory consequence, residual liquids with the observed multi-phase cotectic character (O'Hara *et al.* 1970, 1974; Biggar *et al.* 1971, 1972).

Such an interpretation demands the existence of large volumes of plagioclase-bearing cumulates beneath the maria. Other chemical features of the rocks suggest that a metal phase may also have been fractionated, and unknown amounts of olivine and calcium-poor pyroxene may have been removed. Until these fractionations can be quantified and restored, the composition of the parental magma emanating from the lunar interior cannot be estimated.

As shown above it is no more than an academic exercise to attempt to reason from rocks composition A to an estimate of the lunar mantle composition. It is, however, no easier to reason from liquid B to the lunar mantle composition, than it would be to reason to the terrestrial mantle composition from a late residual liquid of the Bushveld complex, without knowledge of the cumulate sequence in that body.

In conclusion, it is recalled that all arguments based upon the chemistry of chromium, vanadium, barium, rubidium, strontium, etc. have been fully integrated (O'Hara *et al.* 1974) with (and in many cases provide strong support for) the above interpretation of lava flows from the lunar maria surfaces.

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