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Sm-Nd age of lherzolite 67667: implications for the processes involved in lunar crustal formation

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Pristine samples from the lunar highlands potentially offer important information bearing on the nature of early crustal development on all the terrestrial planets. One apparently unique sample of this group of lunar crustal rocks, the feldspathic lherzolite 67667, was studied utilizing the Sm-Nd radiometric system in an attempt to define its age and the implications of that age for the evolution of the lunar highlands. Data for 67667 precisely define an isochron corresponding to an age of 4.18 ± 0.07 AE. The observed lack of disturbance of the Sm-Nd system of this sample may suggest that this time marks its crystallization at shallow depth in the lunar crust. However, the possibility that this age, as well as those of other highland rocks, indicate the time of their impact-induced excavation from regions deep enough in the lunar crust to allow subsolidus isotopic equilibrium to be produced or maintained between their constituent minerals is also considered. Taken together, bulk rock Sm-Nd data for four "high-Mg" rocks, including 67667, indicate that the chemical characteristics of all their source materials were established 4.33 ± 0.08 AE ago and were intimately associated with the parent materials of KREEP. This finding provides more support for the concept of a large-scale differentiation episode early in lunar history. The possible roles of the crystallization of a global magma ocean, endogenous igneous activity, and of planetesimal impact, in producing the observed geochemical and chronological aspects of lunar highland rocks are discussed.

1. Introduction

Data obtained from analysis of the radiometric systems of rocks formed during early lunar history provide important information in trying to reconstruct the nature of lunar differentiation. It is imperative, however, to take into account all possible factors which may influence the chronological data as well as to consider these data in league with what is known of the physical and chemical conditions which controlled primary lunar differentiation events.

We report here results of a study of the Sm-Nd isotopic systematics of a lunar crustal rock recovered by the Apollo 16 mission, the feldspathic lherzolite 67667. Several interpretations of these data are given and an attempt is made to synthesize the chronologic data for lunar highland rocks and determine whether these data uniquely define a sequence of events in early lunar history or are in fact compatible with more than one model of early lunar evolution.

2. Sample characteristics

67667 is a 7.9-g rake sample recovered at the rim of North Ray crater. Warren and Wasson [1] give its mode as 58% olivine, 21% plagioclase, 15% orthopyroxene, 5% clinopyroxene, and 1% ilmenite. Trace element and mineral chemical data [1] indicate that 67667 has low siderophile element abun-

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dances and falls into the group of "high-Mg" [2] crustal rocks on a plot of Ca content of its plagioclase vs. Mg/(Mg + Fe) of its mafic minerals. This classification is supported by the high Ba content of the 67667 plagioclase [3] although the almost chondritic Sm/Ti [4], higher than chondritic Sc/Sm [5], and the calculated initial $^{87}Sr/^{86}Sr$ [6] indicate similarities with the "ferroan anorthosite" group [7]. The very high Sr content of the 67667 plagioclase [3] distinguishes it from both groups.

67667 is severely cataclastized with varying amounts of its plagioclase converted to maskelynite [3,8]. Most grains in our sample had been rendered cloudy in appearance by this granulation event. Our sample was very friable, and we did not observe any single grain larger than about 0.2 mm after gentle disaggregation, whereas Warren and Wasson [8] noted grain sizes up to 0.8 $\times$ 0.5 mm in their section of 67667, suggesting a reasonably coarse grained parent for the 67667 sample. Although apparently unique in the lunar sample inventory this feldspathic lherzolite composition may be a significant chemical component of highland breccias [1].

We received a 1.25-g chip of 67667 which, after preliminary cleaning, was lightly crushed in a boron-carbide mortar. From the crushed material "whole rock" splits totaling approximately 45 mg were removed. The remainder of the sample was then sieved to collect grains in the 125–60 μm size fraction. Plagioclase was separated from this material by conventional magnetic separation followed by hand picking to remove any remaining contaminants. Separation of olivine, clinopyroxene and orthopyroxene proved more difficult. Due, in part, to the effect of the shock granulation of the samples, the distinction in color between these minerals was minimal. Several more steps of magnetic and density separation using filtered methylene iodide were performed to enhance the abundance of the various minerals in different fractions. Although uncertain, it is believed that the olivine content of the two measured pyroxene fractions was less than about 10%. The presence of olivine in these separates should not affect their Sm/Nd or Nd isotopic composition to any significant degree due to the normally very low REE partitioning into olivine.

3. Results

Analytical techniques employed in this work are essentially identical to those described previously [9,10]. Results for 67667 are given in Table 1 and are plotted on the Sm-Nd isochron diagram of Fig. 1.

The Sm concentration measured for our two total rock splits is about 35% higher than that reported by Warren and Wasson [1] demonstrating that "total rock" measurements of small samples from coarse-grained rocks do not produce representative results that approach the level of precision of the analytical technique. Our data suggest that our sample of 67667 was less feldspathic and contained a larger fraction of clinopyroxene compared to that measured by Warren and Wasson [1].

Using distribution coefficients for Nd and Sm in plagioclase [11] and the Nd and Sm concentration of the plagioclase measured here, a liquid in equilibrium with this mineral is calculated to contain substantially more than 100 ppm Nd. While this calculated Nd concentration is an upper limit to the actual value in the parent liquid of 67667, since the presence of a trapped liquid component has been neglected in the calculation, it is obvious that the minerals of 67667 were not formed as cumulates of a mare basalt type liquid. The Sm/Nd

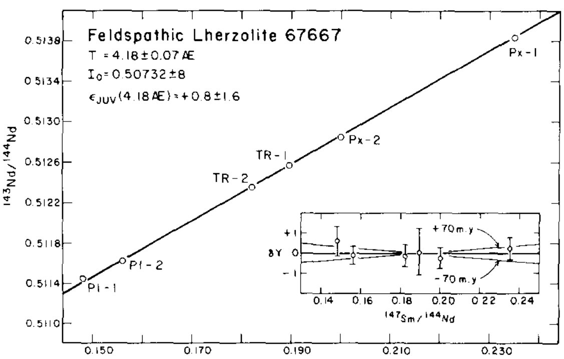

Fig. 1. Sm-Nd isochron diagram for feldspathic lherzolite 67667. Inset shows the relative deviations of $^{143}Nd/^{144}Nd$ for individual data points from the best fit line. Data precisely define a single line indicative of *no* disturbance in the Sm-Nd system of this rock even though its minerals have been highly granulated by shock.

TABLE 1

Analytical results

Sample	Sample weight [a] (mg)	[Sm] (ppm)	[Nd] (ppm)	$^{147}Sm/^{144}Nd$	$^{143}Nd/^{144}Nd$ [b]
Pl-1	5.25	1.751	7.127	0.1481	0.511449
		2	9	2	38
Pl-2	4.82	1.360	5.268	0.1560	0.511631
		1	11	3	22
TR-1	13.85	2.878	9.177	0.1896	0.512570
		4	62	15	59
TR-2	7.57	2.762	9.161	0.1822	0.512356
		2	7	2	27
Px-1	4.85	4.480	11.53	0.2349	0.513834
		3	1	3	28
Px-2	17.34	3.516	10.62	0.2000	0.512843
		1	9	2	25

[a] Sample weight of aliquot used for composition measurement only.

[b] Isotopic data relative to $^{143}Nd/^{144}Nd = 0.511929$ for L.J. Nd standard, and fractionation corrected to $^{148}Nd/^{144}Nd = 0.241572$.

calculated for a liquid in equilibrium with these plagioclase data is close to that measured for the total rock splits, while Sm/Nd calculated for a clinopyroxene in equilibrium with these plagioclases is close to that measured for separate Px-1. The degree of equilibration observed between plagioclase, clinopyroxene, and total rock Sm and Nd abundances suggests that, if 67667 formed as a cumulate, it underwent substantial subsolidus equilibration. In contrast, however, these data may imply that 67667 is not a cumulate, but rather a product of closed system crystallization of an ultramafic magma body in the lunar crust. Regardless of whichever of these two possibilities is correct, it is apparent that the minerals in this rock formed in equilibrium with a liquid which had high REE abundances and a slight relative enrichment in the light REE. A REE enriched parent liquid of this nature is held in common with many members of the "high-Mg" group rocks which have been analyzed [12–15]. At least three inferences can be drawn from this observation:

(1) The "high-Mg" group rocks crystallized from a liquid which had trace element characteristics close to the KREEP component.

(2) Incompatible element-enriched liquids, such as KREEP, were introduced into the source regions of the "high-Mg" rocks as interstitial fluids and allowed to equilibrate or "enrich" already existing materials of the lunar crust. This latter idea is similar to "mantle metasomatism" (e.g. [16]) often invoked to explain incompatible trace element-enriched materials in the earth's mantle, and has been mentioned for the lunar case by Irving [17]. This concept may in some aspects be similar to the hybridization models presented by Hubbard and Minear [18].

(3) Melts from the lunar interior, of either mantle or crustal origin, assimilated substantial quantities of a proto-KREEP material during their ascent towards the surface [19,20]. The assimilated KREEP component would dominate the incompatible element and isotopic characteristics of the magmas but would still allow for the high Mg/(Mg + Fe) of the high-Mg suite magmas.

As shown in Fig. 1, the isotopic data for 67667 precisely define a linear array which indicates an age of 4.18 ± 0.07 AE. The initial (I_0) $^{143}Nd/^{144}Nd = 0.50732 \pm 8$ given by this isochron is within uncertainty the same as the value calculated for the eucrite Juvinas at that time (i.e., ϵ_{JUV} (4.18 AE) $= +0.8 \pm 1.6$ calculated from I_0 alone or $+0.7 \pm 1.0$ using TR-2 combined with uncertainties in the Juvinas data). It is important to note that there is no evidence for a disturbance of the Sm-Nd system, despite the severe granulation of 67667.

4. Age interpretation

Of the ages yet recorded for "pristine" highland rocks of various mineral constitutions, 4.18 AE is among the youngest. The simplest interpretation of this age, especially in light of the well defined isochron, is that it represents the time at which 67667 crystallized from a magma. Whether this magma was residual from an early large-scale differentiation episode (i.e., magma ocean) or was the product of internal melting of the moon at this time will be discussed more fully later. Another interpretation of this age is that it represents the time of excavation of the 67667 material and not its true crystallization age. Because of the lack of any evidence for disturbance in the Sm-Nd data of 67667, and considering the comparatively low degree of shock-induced metamorphism in this sample [3], it is unlikely that the true age of 67667 was "*re*set" by an impact event (e.g. [41]). However, it may be possible that an impact *set* the age of 67667 by the following mechanism. It has now been adequately demonstrated that mineral to mineral Nd isotopic equilibrium is maintained in terrestrial ultramafic xenoliths which have been kept at temperatures $> 1000°C$ prior to their inclusion in a host basalt or kimberlite [21–23], as would be expected from experimental cation diffusion data (e.g. [24]). If such diffusional equilibrium is also maintained or established in lunar materials exposed to sufficiently high temperatures, the radiometric "clocks" in these materials will not begin to record time until such diffusion becomes much slower than changes due to radioactive decay. Excavation from depth is certainly an adequate mechanism to rapidly lower the temperature (after perhaps initial shock heating) and hence initialize a radiometric clock. In this context, it is important to note that the ^{39}Ar-^{40}Ar data of Maurer et al. [25] on Apollo 16 breccias show one group of samples with ages in the range 4.12–4.21 AE. These authors interpreted this time period as marking a time of medium (up to a few hundred kilometers) size cratering, whereas Schaeffer and Husain [26] and Wetherill [27] suggest that it may mark the formation of the Nectaris basin. This latter interpretation may be supported by data obtained from textural studies of the sample collection from North Ray crater [28] which suggest that much of the coarser material, at least at Apollo 16 station 11, may have ultimately been derived from Nectaris ejecta. However, an alternate explanation to the older age group found by Maurer et al. [25] is that the Apollo 16 site is underlain by material, possibly a pluton, which crystallized about 4.18 AE ago and then was rapidly mixed into the regolith by a number of minor impacts. Whichever model is correct, it is interesting that the age recorded by the Sm-Nd system of 67667 also falls within the 4.12–4.21 AE age group found by Maurer et al. [25], and its texture reflects a severe impact.

5. Relation of 67667 to highland chronology

As summarized in more detail in Carlson and Lugmair [15], age data for highland rocks, mainly of the "high-Mg" group, range from 4.18 AE (67667) to 4.45 AE (72417 [29]). Again many of these ages are subject to the question of whether they represent crystallization ages or excavation events. One way to test whether a common time exists when the chemical characteristics of these samples were established is by the construction of a total rock isochron for these materials. Colinearity of total rock data, for a group of chemically related samples such as those of the high Mg suite, indicates that all the samples had identical isotopic compositions at the given time, and that their radioactive parent-daughter ratios were not significantly altered by later events.

Total rock Sm-Nd isotopic data for four highland rocks measured in our laboratory are plotted on the isochron diagram of Fig. 2. As can be seen, these data form a reasonably well defined linear array. The age indicated by these data is 4.33 ± 0.08 AE with an initial $^{143}Nd/^{144}Nd$ essentially equal to that of Juvinas at that time ($\epsilon_{JUV}(4.33)$ AE $= +0.4 \pm 1.8$ using the error from I_0 or 0.4 ± 1.2 with root mean square estimate from I_0 and Juvinas

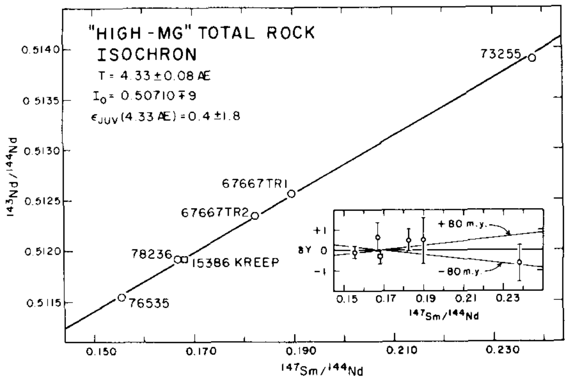

Fig. 2. Whole rock Sm-Nd data for four "high-Mg" lunar highland rocks measured at La Jolla. The alignment of these data about a single line suggests that the parent materials of all these samples had a common Nd isotopic composition 4.33 ± 0.08 AE ago and that very little fractionation of Sm from Nd occurred during the production of the actual samples from their source regions. Data for KREEP basalt 15386, though not included in the calculation of the best fit line, still fall on it within analytical uncertainty suggesting an intimate relationship between the sources of the "high-Mg" highland rocks and KREEP.

data). This age is in remarkable agreement with the average Sm-Nd model age of 4.36 ± 0.06 AE obtained for KREEP-rich materials [10]. This is illustrated by the fact that the total rock Sm-Nd data for the KREEP basalt 15386 fall on the best fit line in Fig. 2. It is also important to note that the Sm-Nd data presented by Oberli et al. [30] for a small number of KREEP-rich samples also produce an average T_{ICE} model age of 4.30 ± 0.16 AE when using the chondritic reference parameters reported by Jacobsen and Wasserburg [31]. The previously reported model ages by Oberli et al. [30], which were in excess of 4.6 AE, apparently resulted from the direct use of the Juvinas Sm-Nd reference parameters reported by Lugmair et al. [14] without taking into account interlaboratory biases in Nd measurements [10].

The coherence of the data in Fig. 2 about a single line provides strong evidence that all of these samples were derived from a common source material about 4.33 AE ago. This is further support for models of a very large-scale early differentiation episode on the moon as reflected in the Sm-Nd, U-Pb, and Rb-Sr systems of a variety of lunar rock types. Furthermore, the near agreement of this 4.33 AE age with that obtained from the model ages of KREEP suggests that the parent material of "high-Mg" highland rocks and KREEP were intimately related. Considering the highly differentiated nature of KREEP-type materials, it appears that the final stages of a primary lunar differentiation were complete by about 4.30 AE ago. However, the occurrence of highland rocks, such as 67667, with ages less than 4.3 AE, shows that significant secondary events also occurred in the lunar highlands.

These pieces of evidence on events occurring during early lunar history can be used to evaluate popular scenarios of the early evolution of the moon. The following evaluation splits the models into two extreme groups, one depicting differentiation through entirely endogenous activity and the other, which describes the effects of outside influence (i.e., collisions). This is done purely for the purpose of discussion and is not meant to imply that events on the moon were solely restricted to either of these possibilities.

6. Primary differentiation

6.1. Endogenous

One popular model of early lunar history calls upon the presence of a global molten layer, up to several hundred kilometers thick, which is allowed to differentiate to produce an anorthositic crust, a deep mafic cumulate zone which later melts to produce the mare basalts, and near the interface, a residual liquid enriched in trace elements which can be considered the parent KREEP magmas. This model has been described extensively [32–34] and will not be explored in depth here. Models of a lunar magma ocean, in their various detailed versions, are capable of satisfying many of the chronological constraints imposed by the various radiometric systems. However, the available radiometric data put only very little constraint on the dimensions of such a "magma ocean". Based on the chemical and radiometric data alone, there does not seem to be a requirement for a global molten zone, only one larger than the area sampled by the various lunar missions. However, the presence of a globe encircling anorthositic crust on the moon may be a most compelling argument for such a global "magma ocean".

6.2. *Impact induced*

The effect of the impact of large planetesimals ($> 10^{22}$ g) during the earliest lunar history is poorly understood. As discussed by Wetherill [27], collision of a large planetesimal, especially into a moon that is already "hot" and hence also quite plastic, may be capable of producing a considerable amount of melting [35]. A melt pool formed by this mechanism is required to have dimensions of several thousand kilometers in diameter and depths approaching 400 km to account for the evidence of early differentiation found at all the lunar sample recovery sites. Let us assume, for a hypothetical case, a melt pool of 2500 km diameter of an average depth of 200 km (10^9 km^3). To totally melt this volume of material, and assuming it was originally 100°C below its melting point, would require an energy input of 1.3×10^{34} erg (specific heat $= 1.2 \times 10^7$ erg/g deg, heat of fusion $= 4 \times 10^9$ erg/g). Assuming an impacting body of 4×10^{22} g, traveling at 13 km/s, which by the scaling law of Grieve and Dence [36] is required to produce a 2500-km-diameter basin [27], 40% of the kinetic energy of the impacting body would have to be transferred as thermal input into the impact region. This percentage is less than that estimated by Kaula (50% [37]) for the impact of a body large enough to produce the Imbrium basin. As pointed out by Kaula [37] the larger the impacting body, the greater will be the percentage of impact energy retained as heat. Hence the production of 10^9-km^3 melt pools in the early history of the moon at least seems physically possible, if not probable, and requires consideration in models of early lunar differentiation.

The evolution of such a huge melt pool, while being thermally distinct from that of a global melt zone, conceivably would lead to similar chemical differentiation trends. Hence, the end products of a large melt pool and a "magma ocean" need not be resolvably different. In fact, the suggestion that the "magma ocean" contained relative REE abundances fractionated from the chondritic pattern [38] may be more readily accounted for by the impact melting of previously differentiated lunar crustal material, rather than calling upon non-chondritic primordial REE abundances in the moon.

7. Later highland differentiation

7.1. *Endogenous*

If the conclusion that primary differentiation ended before 4.3 AE ago is valid, highland rocks which give younger ages must have been produced or modified by events occurring after the early episode. One model for this calls upon the generation of the "high-Mg" rock suite by partial melting of the lunar interior. This melting produced plutons of noritic-troctolitic composition which intruded into the lunar crust [19,20,39]. The available radiometric data for highland rocks are entirely compatible with this idea. However, as discussed previously, the isotopic systematics of these rocks require them to have been generated from, or mixed with, materials which were originally formed during the primary differentiation episode. Furthermore, the alignment of the data in Fig. 2 about a single line suggests that fractionation of Sm/Nd of these samples was minimal during events occurring after primary differentiation. Hence, if these highland rocks were formed by partial melting, the limited Sm/Nd fractionation observed suggests that the degree of melting must have been quite large. Alternatively, the possibility that the REE contents of these rocks are dominated by an assimilated KREEP component [19,20], which was introduced either into the source regions of the "high-Mg" rocks prior to melting, or into the magmas during their ascent towards the surface, must also be considered since the Sm-Nd systematics of KREEP-rich samples [10] indicate that the chemical characteristics of the KREEP component were also established roughly 4.3 AE ago.

7.2. *Impact induced*

The possibility that relatively young ages for highland rocks represent the time of excavation of the sample was discussed earlier. As mentioned, the 4.18-AE age of 67667 may represent the time of basin formation and hence deep excavation of the lunar crust. If a long history of subsolidus isotopic equilibration and subsequent impact excavation is to be considered a favored model for the relatively young ages found for some high Mg suite samples (67667; 73255,27,45 [15]; 76535 [14,40]), these rocks, while still reflecting their formation during earlier events in their bulk characteristics (Fig. 2), must have resided at sufficiently deep levels in the lunar crust where high enough temperatures led to continual resetting of their internal, or mineral to mineral, isotopic systematics until the time of their excavation.

8. Summary and conclusions

Sm-Nd isotopic data for the Apollo 16 lherzolite 67667 re-enforce the observation of a large range in apparent ages obtained for "pristine" highland rocks. Whether this range in ages implies continuing igneous activity throughout the first ~ 400 m.y. of lunar history or represents excavation of deep crustal material by large impacts during this time cannot yet be determined unambiguously.

Regardless of the nature of these events, all of the highland samples which have been analyzed, including KREEP-rich materials, indicate involvement in a large-scale differentiation event which appears to have climaxed approximately 4.35 AE ago. The nature of this event is not defined by the radiometric data alone. Models of a global magma layer or a very large ($> 10^9$ km^3) impact-induced melt pool could both satisfy much of the chemical and isotopic constraints supplied by various lunar materials. The distinction between these two scenarios carries important information regarding the thermal evolution of the terrestrial planets. Unfortunately, the definitive answer is not yet apparent and may, in the end, have to wait until materials from regions far distant from the earth-facing lunar basins can be recovered and analyzed.

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