Lu-Hf constraints on the evolution of lunar basalts

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Lu-Hf CONSTRAINTS ON THE EVOLUTION OF LUNAR BASALTS

Hirokazu Fujimaki* and Mitsunobu Tatsumoto

U. S. Geological Survey, Denver

Abstract. Very low Ti basalts and green glass samples from the moon show high Lu/Hf ratios and low Hf concentrations. Low-Ti lunar basalts show high and variable Lu/Hf ratios and higher Hf concentrations, whereas high-Ti lunar basalts show low Lu/Hf ratios and high Hf concentrations. KREEP basalts have constant Lu/Hf ratios and high but variable Hf concentrations. Using the Lu/Hf behavior as a constraint, we propose a model for the mare basalts evolution. This constraint requires extensive crystallization of the primary lunar magma ocean prior to formation of the lunar mare basalts sources and the KREEP basalts. Mare basalts are produced by the melting of the cumulate rocks, and KREEP basalts represent the residual liquid of the magma ocean. The Lu and Hf concentrations and the Hf isotopic data of lunar rocks suggest that assimilation cannot be accepted as a major process to explain the diversity of the mare basalts. The urKREEP hypothesis is also unnecessary. Both high- and low-Ti basalts show enough iron enrichment to be regarded as melting products of the last stage cumulate rocks from the lunar magma ocean. The KREEP basalts are also rich in iron and may be regarded as the final, residual liquid left after the crystallization of the major portion of the primary lunar magma ocean.

Introduction

In the early stages of lunar studies, a simple cumulate-remelting model was proposed for the evolution of the mare basalts [e.g., Smith et al., 1970; Philpotts and Schnetzler, 1970]. This model was generally accepted and was further developed by other workers [e.g., Taylor and Jakes, 1974, 1977; Shih and Schnellfeld, 1976; Taylor, 1976; Drake and Consolmagno, 1976; Papanastassiou et al., 1977; Nyquist et al., 1977]. This cumulate-remelting model, however, has been challenged by Ringwood and his coworkers with their alternative 'primitive model' [e.g., Ringwood, 1970; Ringwood and Essene, 1970a, b]. In response to numerous criticisms of this model [e.g., Taylor and Jakes, 1974; Lugmair et al., 1975; Taylor, 1975], Ringwood and his coworkers further proposed a 'hybridization model' [Ringwood, 1975; Ringwood and Kesson, 1976]. They refined their earlier model and proposed 'sinking pods' in the lunar mantle [Kesson and Ringwood, 1976; Ringwood and Kesson, 1976]. In the subsequent years the hybridization-sinking pods model has gained acceptance over the cumulate-remelting model, because the problems inherent to the cumulate-remelting model seemed serious enough to invalidate it as a working model.

We show here that a cumulate-remelting model best explains the recently acquired data on the Lu-Hf systematics of lunar mare basalts. Our model is first constructed using the Lu and Hf concentration data and it is then further strengthened by the Hf isotopic evidence [Unruh et al., 1984]. We also show that the similarity of MgO/FeO ratios and the Cr2O3 contents between high-Ti and low-Ti basalts, which have been given significance by Ringwood and Green [1975] and Ringwood [1975, 1976], are not important constraints for lunar basalts petrogenesis. Our principal aim in the present paper is to revive the remelting model for further consideration with the powerful constraints of Lu-Hf systematics of lunar basalts.

Lu-Hf Systematics

Basaltic magmas provide information about the petrological, mineralogical, and geochemical characteristics of the mantle sources. Synthetic experimental studies [e.g., Walker et al., 1975; Hodges and Kushiro, 1974], although important, cannot uniquely predict the mare assemblage of minerals in the magma sources. Major element chemistry is also of limited use in determining the mineralogy of the magma sources. Even trace element geochemistry and isotopic studies cannot uniquely determine the modal compositions of the magma sources.

Since the beginning of Apollo 11 studies, the most controversial topic has been the petrogenesis of mare basalts, which is inevitably related to the characterization of the mare basalt magma sources. In the ensuing debates, important constraints for the petrogenesis have been pointed out by many investigators [e.g., Schnetzler and Philpotts, 1971; Taylor et al., 1972; Tera and Wasserburg, 1974; Lugmair, 1975; Lugmair et al., 1975; Nyquist et al., 1976; Papanastassiou et al., 1977; Lugmair and Marti, 1978; Patchett and Tatsumoto, 1981a]. The principal hypothesis of most of these studies was the cumulate-remelting model. In opposition to the several varieties of remelting models, hybridization models have been proposed by Hubbard and Mines [1976], Kesson and Ringwood [1976], and Ringwood and Kesson [1976]. Ringwood and Kesson [1976] proposed a hybridization model in which ilmenite-pyroxenite 'pods' formed between the lunar crust and mantle during the lunar magma ocean differentiation. The pods that were late-stage cumulate-enriched in Fe, Ti, and incompatible elements sank into the primordial interior and reacted with a large surrounding envelope to produce the variety of lunar basalts.

*Also at Department of Chemistry and Geochemistry, Colorado School of Mines.
depending on the hybridization ratios of the pods and primitive interior material. The hybridization model appears to overcome the three major problems: the heat source, the lack of significant difference in Mg/Fe between the low-Ti basalt and the high-Ti basalt groups, and the lack of difference of Cr abundances between these groups.

Nyquist et al. [1979, 1981] have shown that the REE and Rb-Sr and Sm-Nd isotopic characteristics of the mare basalt were consistent with an equilibrium, modal remelting model but that the magma ocean was light in REE enrichment 15-NK elemental fractionation can be expected to be as large as that between Lu and Nd (or at least Sm), and its effects should be imprinted on both cumulate rocks and their residual liquids. Thus the Lu-Hf fractionation effects should provide clear evidence for the selection of an appropriate model. The Lu-Hf characteristics of the lunar basalts predicted by the ‘sinking pods assimilation model’ [Kesson and Ringwood, 1976; Ringwood and Kesson, 1976]. Because both Lu and Hf are refractory elements, their abundances in the initial lunar magma ocean must be chondritic or perhaps twice chondritic, if the entire moon geochemically simulates the terrestrial mantle after the separation of the earth’s core [e.g., Ringwood and Essene, 1970a]. In this paper, although higher initial Lu and Hf abundances in the moon are not ruled out, we use the chondritic Lu-Hf abundances [Patchett and Tatsumoto, 1981b] as a starting point, because we would like to explain lunar basalt evolution with the help of a minimum number of assumptions.

Lu-Hf Characteristics of the Lunar Mare Basalts

The Lu-Hf characteristics for the lunar mare and KREEP basalts are shown in Figure 1. The data (from literature; see Figure 1 caption) do not include soil and breccia samples. Only green glass samples that are generally thought to be volcanic are included. The chondritic Lu/Hf abundance ratio [Patchett and Tatsumoto, 1981b] is marked by ‘C’ in Figure 1.

Each group of lunar basalts occupies a distinctive region in Figure 1. The very low Ti (VLT) basalts and the green glasses show high Lu/Hf ratios and low Lu and Hf concentrations and their Hf concentrations tend to increase as the Lu/Hf ratio decreases. The Apollo 15 low-Ti basalts are slightly enriched in Lu and show lower Lu/Hf ratios. In contrast, the Apollo 12 low-Ti basalts have Lu/Hf ratios as high as some of the VLT basalts, and the Hf concentrations in these basalts are slightly higher than Apollo 15 basalts. These basalts seem to lie on the extension of the high-Ti, low-K basalt area. The Lu/Hf ratios of the Apollo 15 low-Ti basalts tend to decrease with increasing Hf concentrations and tend to reach the high-Ti, low-K basalt region. Also, the distribution appears to be continuous from the high-Ti, low-K basalt region to the high-Ti, high-K basalt area. The Lu/Hf ratios of the high-Ti, low-K basalts seem to be either constant or tend to increase slightly with increasing Hf concentrations. In contrast, the Lu/Hf ratios of the high-Ti, high-K basalts decrease slightly with increasing Hf concentrations. The KREEP basalts are characterized by almost constant Lu/Hf ratios and high Hf abundances, and there is some overlap with the high-Ti and high-K basalt field. The boundary among the basalts groups is clear and there is some degree of overlap. This overlap may be caused by either analytical error or by the apparent lack of homogeneity due to the extremely small sample-size analyzed.

These Lu-Hf data, in combination with partitioning data and Hf isotopes, can provide some constraints on magma production and genetic relationships among the various basalts. First, let us consider the most primitive green glasses and the VLT basalts. Provided the sources are not ilmenite-rich, the Lu/Hf ratios of the basalts should be lower than those of their sources, because $DLu > D_H$, and it is usually much larger than that for Hf. On the other hand, the sources of the Apollo 12 low-Ti basalts and green glasses should have had very high Lu/Hf ratios, certainly much higher than the chondritic ratio. This means that even green glass samples and the VLT basalts cannot be direct derivatives of a chondritic or primitive material that is alleged to exist in the lunar interior [e.g., Kesson and Ringwood, 1976; Ringwood and Kesson, 1976]. These green glass samples should have been derived from some differentiated material, such as rocks accumulated from the lunar magma ocean.

The source material of the Apollo 12 low-Ti basalts should also have had a higher Lu/Hf ratio than the chondritic ratio. This material would be expected to have been more enriched in Hf. On the other hand, the sources of the Apollo 15 low-Ti basalts could have had slightly lower Lu/Hf ratios and higher Hf concentrations than those of the Apollo 12 basalts. Although the distribution of the high-Ti, low-K basalts seems to be continuous with that from the low-Di 12 low-Ti basalts, the parental sources of the high-Ti, low-K basalts could have had Lu/Hf ratios similar to the chondritic value. Their parental sources should have contained enough ilmenite to explain the high-Ti contents of the
Fujimaki and Tatsumoto: Lu-Hf Constraints

Fig. 1. Lu-Hf features for VLT and green glass (●), Apollo 12 low Ti basalts (○), Apollo 15 low-Ti basalts (●), high-Ti and low-K basalts (●), high-Ti and high-K basalts (●), and KREEP (■). Points with (□) symbols are accompanied with Hf isotopic data. The dotted line represents the Lu/Hf-Hf trend of the magma ocean (see Figure 3) and starts from 0.17 ppm Hf and Lu/Hf ratio 0.2235, which are chondritic Lu and Hf abundances reported by Patchett and Tatsumoto [1981b]. The data are from Wanke et al. [1970, 1974, 1976]; Nakata et al. [1970, 1971]; Morrison et al. [1970, 1971]; Haskin et al. [1970]; Kharker and Turekian [1971]; Taylor et al. [1971]; Brunfelt et al. [1971, 1974]; Boynton et al. [1976]; Ma et al. [1976, b, 1980, 1981]; Laul and Schmitt [1973, 1974]; Laul et al. [1978]; Lindstrom et al. [1976, 1977]; Rhodes et al. [1976, 1977]; Warner et al. [1979, 1980]; Beatty et al. [1979]; Wentworth et al. [1979]; and Unruh et al. [1984].

Basalts and should have been enriched in incompatible elements. Some high-Ti, low-K basalts have relatively high Lu/Hf ratios and they seem to plot in an extension of the high-Ti, high-K basalt field. However, because of the scarcity of the data, it is not clear whether they are really part of the high-Ti, high-K basalt group. The source materials of the high-Ti, high-K basalts could have had a near-chondritic Lu/Hf ratio. Although the high-Ti, low-K basalts and the high-Ti, high-K basalts have almost the same Ti abundances, their Hf concentrations as well as those of other incompatible elements vary by more than a factor of two.

It is very difficult to produce the observed Lu-Hf distributions of the KREEP basalts by cumulate remelting. Their variation in Hf abundances is so large that, if they were formed by cumulate remelting, an extremely large range of partial melt percentage or a large range in Hf concentration in the sources is required to produce this distribution. However, the Lu/Hf ratios of KREEP basalts are nearly constant. This constancy of Lu/Hf ratios requires that the Lu partition coefficient between their magma sources and KREEP had to be the same as that of Hf, and also that the source rocks are required to have had the same Lu/Hf ratios as the KREEP.

Lu-Hf Evolution Model

Assumption 1: Modal Mineralogy and Lu, Hf Abundances of the Moon

Although the best way to make a model is with few assumptions, we have to characterize the mineralogy of the entire moon on the basis of experimental results [e.g., Kushiro et al., 1971; Hodges and Kushiro, 1974; Walker et al., 1972, 1971, 1975] and the modal mineral composition of the whole moon is inferred from estimated chemical compositions of the moon [Ganapathy and Anders, 1974; Taylor and Jakos, 1974; Taylor, 1976; Ringwood and Kesson, 1976]. However, these estimates vary considerably because pressure, temperature, and extremely low fO2 complicate the phase relations to such a degree that the modal mineralogy of the moon cannot be uniquely determined. Therefore we simply assume from various analogies that the whole-moon consists of 65% olivine, 15% orthopyroxene, 9.9% clinopyroxene, 0.1% ilmenite, and 10% plagioclase. Spinel is ignored because it does not affect the Lu-Hf systematics. We accept chondritic Lu and Hf elemental abundances for the entire moon as determined by Patchett and Tatsumoto [1981b].
Fig. 2. The relationships between the degree of solidification and crystallizing minerals. Degree of solidification is shown with percent. The numbers above the figure indicate the degree of solidification when each mineral reaches the liquidus line.

**Assumption 2: Crystallization Sequence**

Experimental petrology has helped to clarify the crystallization sequence of minerals in the lunar magma ocean [e.g., Hodges and Kushiro, 1974; Walker et al., 1972]. Although these experimental results cannot quantitatively predict the modal mineralogy of the lunar mantle, we have to assume a quantitative crystallization sequence for the following discussion. Assuming that the initial lunar magma ocean was 500 km thick, olivine should be the first liquidus phase, followed by orthopyroxene, clinopyroxene, plagioclase [e.g., Hodges and Kushiro, 1974; Walker et al., 1972] and finally ilmenite. Two crystallization modes can be considered: one is perfect equilibrium crystallization and the other is maximum fractional crystallization. Although these are the simplest crystallization modes, both are extremes. The actual crystallization that took place is probably between the two extremes. In addition, the modal proportions of the precipitating minerals from the magma should vary gradually throughout the course of solidification of the magma. Therefore the instantaneous amounts of crystallizing minerals are expressed by linear functions for olivine and plagioclase, assuming that olivine and plagioclase continue to crystallize from the beginning to the end, and expressed by parabolic functions for orthopyroxene, clinopyroxene, and ilmenite as follows:

\[
\begin{align*}
\text{o} & \quad f(x) = -1.3 \ x + 1.3, \quad 0 \leq x \leq 1 \\
\text{p} & \quad f(x) = -0.2 \ x + 0.2, \quad 0 \leq x \leq 1 \\
\text{opx} & \quad f(x) = -1.7578115(x - 0.2)(x - 1), 0.2 \leq x \leq 1 \\
\text{cpx} & \quad f(x) = -22(x - 0.7)(x - 1), 0.7 \leq x \leq 1 \\
\text{i} & \quad f(x) = -48(x - 0.92)(x - 0.97), 0.92 \leq x \leq 0.97
\end{align*}
\]

Where the initial amount of the lunar magma ocean is 1, the total amount of each mineral is:

\[
\begin{align*}
\text{o} & \quad \int_0^1 f(x) \, dx = 0.65 \\
\text{p} & \quad \int_0^1 f(x) \, dx = 0.10 \\
\text{opx} & \quad \int_{0.2}^1 f(x) \, dx = 0.15 \\
\text{cpx} & \quad \int_{0.7}^1 f(x) \, dx = 0.099 \\
\text{i} & \quad \int_{0.92}^1 f(x) \, dx = 0.001
\end{align*}
\]

The variable \( x \) is simply numerical and does not mean the degree of solidification. The degree of solidification (\( F \)) can be obtained by

\[
\sum f(x) = F
\]

Our numerical calculation using the above equations show that orthopyroxene reaches the liquidus line after 27% solidification and clinopyroxene appears as a liquidus phase after 78.5% solidification and both continue to crystallize to the end. Ilmenite begins to crystallize after 97.3% solidification. These crystallization sequences are shown in Figure 2. Although this calculation method is purely numerical, the modal assemblages of precipitating rocks can be gradually changed and, according to this change, the partition bulk coefficients can be varied.

Using mass balance, a trace element concentration in liquid can be expressed as follows:

\[
C_e^n = \sum \left( C_e^0 \int_0^1 f(x) dN + \int_0^1 C_e \int_0^1 f(x) dN \right) dx dN
\]

where \( C_e^0 \) is the concentration of element \( e \) in a liquid, \( d \) is the partition coefficient of element \( e \) between phase \( i \) and the liquid, and \( d \) is an integral span, which is variable. Although \( \chi \) is simply a numerical variable, \( \chi \) is controlled by the integral span \( d \). Changing \( d \), we can control the character of crystallization. For example, when \( d \) is large, the crystallization is close to perfect equilibrium crystallization, and when \( d \) is small, the crystallization has characteristics close to the maximum fractionation.

We computed the variations of Lu and Hf abundances for the successive fractionation of the magma ocean and for the cumulate rocks at various degrees of solidification using \( d = 0.001 \) for early to late-stage and \( d = 0.0001 \) for the latest stage during which ilmenite begins to crystallize and finally disappears before total solidification. During the early to late-stage, \( \sim 0.1\% - \sim 0.15\% \) of the whole magma ocean is solidified in each step, and in the latest stage, \( \sim 0.001\% - \sim 0.005\% \) of the whole magma ocean is solidified in each step. Although each step is a
TABLE 1. Partition Coefficients of Lu and Hf Used in the Calculation

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<th>Mineral</th>
<th>Olivine</th>
<th>Orthopyroxene</th>
<th>Clinopyroxene</th>
<th>Plagioclase</th>
<th>Ilmenite</th>
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<td>0.400</td>
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<tr>
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<td>0.140</td>
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All the partition coefficient data are estimated from Fujimaki et al. [1984].

perfect equilibrium crystallization as a whole, this calculation introduces some characteristics of maximum fractionation. We believe that this approach is more realistic than either perfect equilibrium crystallization or maximum fractionation. One tenth of one percent of the initial magma ocean is sufficient to cover the entire lunar surface circa 400 m-thick, even if we assume a depth of only 500 km for the initial magma ocean [e.g., Taylor, 1978]. In this calculation, plagioclase is assumed to float before clinopyroxene begins to crystallize, and after that plagioclase is included in the cumulate rocks. The partition coefficient data that we have used in these calculations are shown in Table 1 and are taken from Fujimaki et al. [1984]. We did not change the partition coefficients for any minerals throughout the course of the solidification.

Results

Our results are summarized in Table 2 and Figure 3. The early to late stage cumulate rocks are extremely depleted in both Lu and Hf. After orthopyroxene begins to crystallize, the Lu/Hf ratios of the cumulate rocks are much higher than those of dunite cumulates because $D_{\text{Lu}}/D_{\text{Hf}}$ is very large [Fujimaki et al., 1984]. In the late stage, after clinopyroxene reaches the liquidus, the Lu/Hf ratios of the cumulate rocks become slightly lower, because $D_{\text{Ppx}}/D_{\text{Cpx}}$ is smaller than that of orthopyroxene. After ilmenite appears as the liquidus phase, the Lu/Hf ratios of the cumulate rocks become low, for even a small amount of ilmenite can be effective in reducing the Lu/Hf ratios of the cumulate rocks. However, after the amount of crystallizing ilmenite once reaches a maximum, the amount of ilmenite begins to decrease, which results in increasing Lu/Hf ratios of the last cumulates. The Lu/Hf ratio in the lunar magma ocean gradually decreases during the early to late stage, and during later stages the ratios seem nearly constant but still decrease slightly.

In Figure 3, some coexisting liquids and cumulate rocks are tied with dashed lines. Before KREEP liquids can be produced, ilmenite-rich cumulate rocks are formed. The cumulate rocks enriched in ilmenite have important characteristics. Relatively early ilmenite-enriched cumulate rocks contain 0.5 to 2 ppm Hf and later ilmenite-enriched cumulate rocks contain 2 to 3 ppm Hf. Their Lu concentrations, and presumably those of other incompatible elements, also differ approximately by a factor of two. On the basis of the Lu-Hf characteristics of the lunar basalts and our calculations described above, we propose the following model of evolution for the lunar mare basalts.

Mare Basalt Petrogenesis

We have calculated the variation of the Lu and Hf abundances in the liquids produced by modal partial melting of the cumulate rocks for a variety of lunar basalts. Clinopyroxene-dominated partial melting is needed only for the Apollo 15 low-Ti basalts. These results are shown in Figure 4. Our calculations clearly demonstrate that 5% to 10% partial melting of the later ilmenite-rich cumulate rocks can produce the Lu-Hf distribution of the high-Ti, high-K basalts, and that 5% to 20% partial melting of the early ilmenite-rich cumulate rocks can produce the high-Ti, low-K basalts. Unruh et al. [1984] also concluded from REE, Nd, and Hf isotope data that the mare basalts were derived by small degrees (<<10%) of partial melting of cumulates sources. The Apollo 12 low-Ti basalts can also be produced by various degrees (less than 30%) of partial melting of ilmenite-poor cumulate rocks. The Lu-Hf features of the VLT basalts and green glasses can be reproduced in our calculation by 5% to 30% partial melting of the ilmenite-free, but late-stage, cumulate rocks. The Apollo 15 low-Ti basalts, however, cannot be accounted for in our calculation by modal partial melting. Most of the Apollo 15 low-Ti basalts plot below the magma ocean trend of Lu and Hf variation. As long as we employ a modal partial melting model, the resulting liquids never cross the magma ocean trend. However, non-modal partial melting dominated by clinopyroxene can explain the Lu-Hf distribution of the Apollo 15 basalt.

Some important points of our model should be emphasized, especially with regard to the ilmenite-rich cumulates. The early ilmenite-rich cumulate rocks contain 0.5 to 2 ppm Hf and they are expected to be enriched by similar factors in other incompatible elements. The later stage ilmenite-rich cumulate rocks contain the same amount of ilmenite, but 2 to 3 ppm Hf and 12 to 18 times chondritic incompatible trace element abundances. The former cumulate rocks can produce the high-Ti, low-K basalts, which should be somewhat enriched in incompatible elements. The latter cumulate rocks can produce the high-Ti, low-K basalts that are highly enriched in incompatible elements. Both sources should produce basalts with almost the same TiO2.
### TABLE 2. Representative Calculated Results of Interest for Lunar Magma Ocean and Cumulate Rock

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(1) Degree of solidification of the lunar magma ocean; (2) Percentage of olivine crystallized at the designated F$_2$; (3) Percentage of orthopyroxene crystallized at the designated F$_2$; (4) Percentage of clinopyroxene crystallized at the designated F$_2$; (5) Percentage of plagioclase crystallized at the designated F$_2$; (6) Percentage of ilmenite crystallized at the designated F$_2$; (7) Lu concentration (ppm) in cumulate rock from the magma ocean; (8) Hf concentration (ppm) in cumulate rock from the magma ocean; (9) Lu concentration (ppm) in successive liquid of the magma ocean; (10) Hf concentration (ppm) in successive liquid of the magma ocean; (11) Lu/Hf ratio of the cumulate rock; (12) Lu/Hf ratio of the successive liquid. *Plagioclase is assumed to float before it is 80% solidified. Lu and Hf concentrations in plagioclase are not added to the cumulate rock before it is 80% solidified.
Fujimaki and Tatsumoto: Lu-Hf Constraints

Fig. 3. Lu-Hf variations of the lunar magma ocean and the complementary cumulate rocks. Coexisting liquid and cumulate rocks are tied with dashed lines. The points of solidification (F; Table 2) are: a = 80%, b = 96%, c = 97%, d = 97.5%, e = 98%, f = 98.5%, g = 99%, and h = 99.5%. The area with a dotted line indicates KREEP.

Contents, but they should have quite different incompatible element abundances. These features exactly match the observations. However, this model does not require the clear distinction between the two basalt groups as shown in Figure 1. We predict that if sampling of the lunar basalt were extended to the entire lunar surface, the boundary between these groups might disappear. Since TiO$_2$ contents of both the high-Ti basalt groups are rather high, even 6% ilmenite in the source cannot account for the high TiO$_2$ contents in these basalts, and we are forced to assume that some of the TiO$_2$ in the source was in clinopyroxene. Other ways to explain the high Ti contents are either by nonmodal melting or by changing the Hf partition coefficient between ilmenite or clinopyroxene and liquid. In the case of nonmodal melting, increasing the contribution from ilmenite to the liquid can easily account for the Ti contents of the basalts. If the Hf partition coefficient is much lower than 1.8, for example 1 or 0.9, we can precipitate more ilmenite (up to 10% to 12%) in the cumulate rocks. Combining the expected Ti contribution from clinopyroxene, the same Lu-Hf distribution and high-Ti content for both groups

Fig. 4. Lu-Hf relationships of the liquids produced by partial melting of the cumulate rocks. Solid circles are cumulate rocks, solid triangles indicate the Hf concentrations and Lu/Hf ratios of the liquids produced by 5% modal partial melting of the corresponding cumulate rocks. Lu-Hf variations according to the degree of modal melting are shown by asterisks (10% to 30%, for example). Open triangles indicate the liquid produced by non-modal melting dominated by clinopyroxene. Each rock type is indicated by a dotted circle.
basalts is not precisely defined, they have LREE- were mixed with the more evolved residual liquid
urKREEP in the lunar interior becomes basalt samples analyzed by Unruh et al. [1984]
unnecessary. have low Lu/Hf ratios and relatively low Hf
magma ocean trend. In Figure 5b, the variations of TiO\textsubscript{2} contents of
melt rocks. The line is the Lu-Hf variation of can change slightly. Therefo Spme data plotted
enriched patterns that are almost paral!el to or partial melts derived from more evolved
Although the boundary between KREEP and'KREEPy parental sources had high Lu/Hf ratios. If they
exactly. Therefore the existence of hypothetical for by hybridization. The Apollo 12 low-Ti
ocean (Figure 3)match the KREEP Lu-Hf variations Hf isotopic differences are not easily accounted
Wasson et al. [1977]. The Lu-Hf variations are taken from Unruh et al. [1984]. The Hf
Fig. 5. (a) The Lu-Hf distributions for KREEP with meteoritic components'; The samples that
maintain DLu/DHf = 1. Otlerwise it is impossible enriched characteristics, even the l'east evolved
sample has almost the same Hf concentration as isotopic ratios (176Hf/177Hf) and Hf
of KREEP basalt (Figure 1). The Lu/Hf ratios of these two samples can represent the real liquids
samples available is small, the observed chemical variation probably encompasses the complete range of
KREEP basalt (Figure 1). The Lu/Hf ratios of the KREEP basalts show as little a variation as their Sm/Nd ratios reported by Lugmair and
Carlson [1978].

Duncan et al. [1976] have reached the conclusion that 'KREEP or its source region have previously undergone ilmenite fractionation and removal.' If KREEP basalts were produced by partial melting of cumulate rocks [e.g., Walker et al., 1972; Weil and McKay, 1975] their sources had to contain enough ilmenite to maintain \( \frac{D_{\text{Lu}}}{D_{\text{Hf}}} = 1 \). Otherwise it is impossible to produce a complete spectrum of KREEP basalts having almost constant Lu/Hf ratios irrespective of their Hf concentrations. We reject the remelting model for the origin of KREEP basalts for this reason.

The Hf abundance of the most enriched KREEP sample is quite similar to that of the estimated 'pure' KREEP [e.g., Laul and Schmitt, 1973; Warren and Wasson, 1979]. The least enriched sample has almost the same Hf concentration as the most enriched KREEP sample reported by Wasson et al. [1977]. The Lu-Hf variations calculated for the final 1% of the lunar magma ocean (Figure 3) match the KREEP Lu-Hf variations exactly. Therefore the existence of hypothetical urKREEP in the lunar interior becomes unnecessary.

Some KREEPy highland basalt samples were returned by Apollo missions 14, 15, 16, and 17. Although the boundary between KREEP and KREEPy basalts is not precisely defined, they have LREE-enriched patterns that are almost parallel to each other [e.g., Drake et al., 1973; Haskin et al., 1973; Wasson et al., 1977].

In Figure 5a, the Lu-HF features of the KREEPy basalts and KREEP-rich clastic samples [e.g., Drake et al., 1973; Haskin et al., 1973; Wasson et al., 1977] are shown along with KREEP data. We obviously omitted some anorthositic samples. The number of the data are limited and most of the samples may not represent their parental magma. Nevertheless, the Lu-HF distribution of the KREEPy basalts can be plotted along the lunar magma ocean trend. This distribution may still contain anorthositic samples, samples assimilated with anorthositic materials, or samples mixed with meteoritic components. The samples that underwent assimilation or mixing show decrease in their Hf concentrations and their Lu/Hf ratios can change slightly. Therefore some data plotted far below or above the magma ocean trend can be considered to have been along the trend before they suffered assimilation or mixing processes. In Figure 5b, the variations of TiO\textsubscript{2} contents of these samples are shown. The dashed line is presumed TiO\textsubscript{2} variation for the lunar magma ocean. Combining the Lu-Hf variation and TiO\textsubscript{2} of both the KREEP basalts and the KREEPy basalts, it seems that the TiO\textsubscript{2} contents of the KREEPy basalts have no relationship with their Hf concentrations. This may also be caused by an assimilation process as proposed by Warren and Wasson [1979] or by meteorite bombardments. However, two samples show the highest TiO\textsubscript{2} concentrations around 10-15 ppm Hf. According to our model, the highest TiO\textsubscript{2} content for the residual liquid of the magma ocean can be expected around 10-20 ppm Hf from which the ilmenite-rich cumulate rocks precipitated. If these two samples can represent the real liquids of the magma ocean, the relationship between Hf and TiO\textsubscript{2} is consistent with our model. The liquids from which ilmenite-free cumulate rocks could have precipitated should not be so enriched in TiO\textsubscript{2}. On the contrary, the liquids that coexisted with ilmenite-rich cumulate rocks should be more enriched in TiO\textsubscript{2}. After ilmenite has been settled out, the TiO\textsubscript{2} contents of the residual liquids should decrease. We emphasize that, because of their KREEP-enriched characteristics, even the least evolved KREEPy samples (whose chemical characteristics have already largely changed) may have experienced at least 80% to 90% fractional crystallization from the presumed initial magma ocean.

**HF Isotopic Evidence**

The relationships between initial-Hf isotopic ratios (\( ^{176}\text{Hf}/^{177}\text{Hf} \)) and Hf concentrations are shown in Figure 6. These data are taken from Unruh et al. [1984]. The Hf isotopic data are shown for KREEP and KREEPy samples. The HF isotopic differences are not easily accounted for by hybridization. The Apollo 12 low-Ti basalt samples analyzed by Unruh et al. [1984] have low Lu/Hf ratios and relatively low Hf contents (Figures 1, 3, and 4), but they have very high Hf values, which means that their parental sources had very high Hf ratios if they were mixed with the more evolved residual liquid or partial melts derived from more evolved
cumulate rocks, then both their Lu/Hf ratios and ε_Hf values would be low. The Hf isotopic evidence show that at least the low-Ti basalt sources had very high Lu/Hf ratios. Although no data is available for the VLT basalts and green glass samples, their ε_Hf can be expected to be much higher than those of the low-Ti basalts if the model holds.

The high-Ti basalts have low ε_Hf values, which are consistent with low Lu/Hf ratios in their parental sources. They cannot come from the same source as the low-Ti basalts. We emphasize that although the high-Ti, low-K basin sources and the high-Ti, high-K basin sources had similar Lu/Hf ratios, the latter sources were approximately twice as enriched in incompatible elements. Therefore these two sources probably formed within a very small range of solidification of the magma ocean: one is enriched in incompatible and the other more enriched, but they have similar degrees of solidification [e.g., Neumann et al., 1954; McIntire, 1963] as evidenced by the similar Lu/Hf ratios in their sources. Our conclusion is that only the last stage of crystallization can make these cumulates.

Mg/Fe and Cr

Ringwood and his associates [e.g., Ringwood and Green, 1973; Ringwood, 1975; Ringwood and Kesson, 1976] noted that the similarities in Mg/(Mg + Fe) ratios and Cr_2O_3 contents between low-Ti and high-Ti basalt sources cannot be satisfactorily accounted for by remelting of olivine + pyroxene + ilmenite cumulates formed during lunar primary differentiation. However, we do not believe that these similarities negate our model, even though we use an extreme fractionation model to produce the lunar mare basalt source. Crystallization is assumed to be a natural process, nor can perfect equilibrium crystallization be achieved throughout the course of differentiation. However, we do not believe that these similarities negate our model, even though we use an extreme fractionation model to produce the lunar mare basalt source.

Most of the mare basalts contain olivine phenocrysts [e.g., Keil et al., 1971; Papike et al., 1976], and even some KREEP-rich basalts contain olivine phenocrysts [e.g., Drake et al., 1973; Wasson et al., 1977]. In addition, experimental studies [e.g., Walker et al., 1972] suggest that olivine can coexist with a liquid at most of the pseudoperitectic point of the SiO_2-olivine-anorthite pseudobinary system. Therefore we can use the Mg-Fe partitioning between olivine and liquid to estimate how the MgO/FeO ratio of the lunar magma ocean could have changed throughout the course of differentiation.

Roeder and Emslie [1970] determined Mg-Fe partitioning between olivine and liquid. They have shown that the Mg-Fe partitioning between olivine and liquid is ~0.30, and that K_D is independent of temperature. They have also shown that K_D is constant at all Ti, the molar FeO/MgO ratio of the liquid is about 1. Because Fe^3+ is not expected in the lunar mantle or in lunar magma [e.g., Sato et al., 1973], these Mg-Fe relationships are applicable to the lunar data.

In Figure 7, the molar MgO/FeO ratio of the lunar primordial interior [Ringwood and Kesson, 1971] is approximately close to the initial magma ocean. According to the Mg-Fe partitioning, the molar FeO/MgO ratio of olivine (point D) and the liquid of the initial magma ocean (point C) with which the olivine coexists are shown in Figure 7. The olivine is approximately Fo 91, and if maximum fractional crystallization is assumed the successive liquid becomes extremely iron-rich, and the chemical composition of olivine crystallized from magma ocean should encompass Fo 96 to fayalite. However, this is unrealistic because maximum fractional crystallization cannot be a natural process, nor can perfect equilibrium crystallization be achieved throughout the course of solidification, olivine that crystallized from the final liquid (point E) should have a molecular FeO/MgO ratio (point F, Fo 89) very close to the initial magma ocean. The molar FeO/MgO ratio of the final and residual liquid coexisting with the olivine is approximately Fo 96 to 0.72. These relationships are shown schematically in Figure 8 [Bowen and Schairer, 1935]. In Figure 8, when the maximum fractional crystallization is attained, the MgO/FeO ratios of final olivine and final liquid should be at point B, and when perfect equilibrium crystallization is accomplished, the MgO/FeO ratios of final olivine and final residual liquid are at points F and E, respectively. Therefore only one quantitative constraint can be inferred from Figure 8. Because natural fractionation processes are between both extremes, the final residual liquid of the lunar magma ocean must have an MgO/FeO ratio somewhere between points E and B in Figure 8.

KREEP and some KREEPy basalts data [e.g., Drake et al., 1973; Wanke et al., 1976; Lindstrom et al., 1977; Warren and Wasson, 1979] are plotted in Figures 7 and 8. All the KREEP and KREEPy basalts and estimates for KREEP [Warren and Wasson, 1979] plot between points E and B. These facts again show that the KREEP basalts can be produced by crystallization differentiation from the lunar magma ocean, and that the KREEPy basalts are iron-rich enough to be considered as final residual liquids of the lunar magma ocean. The ranges of MgO/FeO ratios for the VLT basalts and green glasses, low-Ti basalts, high-

![Figure 6](image-url)

**Fig. 6.** Hf isotopic data versus Hf concentration. Symbols are the same as those in Figure 1. 176Hf/177Hf ratios are corrected for Lu decay. Data are from Unruh et al., [1984].
Fig. 7. FeO-MgO relations between liquid and presumed coexisting olivine. The FeO/MgO ratios are molar. The line A→B indicates an equilibrium relation between olivine and liquid \( K_D = 0.3 \), reported by Roeder and Emslie [1970]. C: FeO/MgO ratio of the initial magma ocean. D: FeO/MgO ratio of olivine, which first crystallizes from the magma ocean. E: FeO/MgO ratio of the final and residual liquid when a perfect equilibrium is accomplished. F: FeO/MgO ratio of olivine, which can coexist with the final liquid, E. Approximate ranges occupied by the basalt groups are shown with FeO/MgO ratios of presumed coexisting olivine.

Although the low-Ti and high-Ti basalts are too iron-rich to estimate the composition of the coexisting olivine in their source, the MgO/FeO ratios of olivine of the presumed source rocks which coexist with the liquids are also shown in Figures 7 and 8 [e.g., Agrell et al., 1970; Haramura et al., 1970; Laul and Haggerty, 1974; Wänke et al., 1976; Papke et al., 1981]. There is no significant difference in the FeO/MgO ratio among the low-Ti basalts, the high-Ti basalts, the low-K basalts, and the high-Ti, high-K basalts. The VLT basalts and green glasses, however, have higher MgO/FeO ratios.

The best developed assimilation model is that of Ringwood and Kesson [1976; Ringwood and Kesson, 1976]. To explain the Lu-Hf systematics using their model, we must argue against the hypothesis of assimilation.

Similar Cr contents for the low-Ti and high-Ti mare basalts can also be predicted by our model. Haggerty et al. [1970] pointed out that lunar olivine contains significant Cr²⁺ and Smith [1974] noted that lunar olivine contains more Cr than terrestrial olivine. However, there is a problem regarding this comparison in that the lunar oxygen fugacity is quite different from the \( F_O^2 \) in the terrestrial mantle. Nevertheless, the lack of significant difference in Cr abundances between the low-Ti and high-Ti basalts can also be explained by our model. Because both high-K basalts, and high-Ti and high-K basalts are also shown in Figures 7 and 8 [e.g., Agrell et al., 1970; Haramura et al., 1970; Laul and Haggerty, 1974; Wänke et al., 1976; Ma et al., 1981]. There is no significant difference in the FeO/MgO ratio among the low-Ti basalts, the high-Ti basalts, the low-K basalts, and the high-Ti, high-K basalts. The VLT basalts and green glasses, however, have higher MgO/FeO ratios.

Sources were formed within a small range of the solidification of the lunar magma ocean, significant differences in Cr contents between the low-Ti and high-Ti sources cannot be expected. However, in a general sense, slightly higher Cr abundances for the VLT basalts and green glasses can be expected. Once again the predictions of our model match the observed characteristics of the basalts.
conclude the following: the VLT basalt and green glass magma sources must have had the highest Lu/Hf ratios, inferred from the high Lu/Hf ratios in the basalts and glasses, and the low Ti basalts magma sources must have had high Lu/Hf ratios, inferred from the high $c_{\text{Hf}}$ values of the low-Ti basalts. The high-Ti basalt sources must have had low Lu/Hf ratios because the high-Ti basalts have low $c_{\text{Hf}}$ and low Lu/Hf ratios. But if we assume that the ilmenite-rich ‘sinking pod’ had a low Lu/Hf ratio and fairly high Hf content, then mixing with the primordial interior (which has low Hf and a chondritic Lu/Hf ratio) cannot produce the initial Hf characteristics of the VLT basalts, green glass, and low-Ti basalts. Even if the ‘sinking pod’ contains a significant amount of trapped liquid [Ringwood and Kesson, 1976], it is still impossible to produce the VLT, green glass, and low-Ti basalts by partial melting of the lowermost part of the primordial interior and the ‘sinking pod’.

Thus there appear to be two choices remaining to save the ‘sinking pod’ hypothesis: one is that the ‘sinking pod’ had a high Lu/Hf ratio but low Hf abundance similar to ilmenite-free cumulate rocks, and the other is that the ‘sinking pod’ itself has a high Lu/Hf ratio, mixed with earlier cumulate rocks with high Hf/Lu ratios. In the former case, because the ‘sinking pod’ is more depleted in Hf and Lu than the primordial interior, mixing with the primordial interior cannot produce basalts with higher Lu/Hf ratios than the low-Ti basalts. Therefore in order to produce the VLT, green glass, and low-Ti basalts, only the ‘sinking pod’ has to melt before it mixes with the primordial interior. In the latter case, only a mixture of very small amounts (<1%) of ilmenite-rich pod rock and large amounts of relatively early cumulate rock can account for the VLT, green glass, and low-Ti basalts. Thus although this process cannot be ruled out completely, it has to be defined as definitely unnecessary. As a major process, at least for the generation of low-Ti and VLT basalts and green glasses.

Summary

The VLT basalts and green glasses have high and variable Lu/Hf ratios and low Hf concentrations, and the low-Ti basalts have lower Lu/Hf ratios and slightly higher Hf concentrations. In contrast, the high-Ti, low-K basalts have low Lu/Hf ratios and are enriched in Hf relative to Lu. The REE patterns of the low-Ti basalts are consistent with the thought that KREEP are residual liquids of the lunar magma ocean. The KREEPy highland basalts may represent the liquid of a highly evolved lunar magma ocean by various degrees of fractionation before assimilation processes or meteorite bombardments complicated their chemistries. The so-called uKREEP hypothesis is unnecessary. We emphasize that the VLT basalt and the green glass samples have very high Lu/Hf ratios, which shows that they were not derived from ‘primordial and chondritic’ material.

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even the VLT or green glass samples are remelting products of a cumulate that formed after at least 80% to 90% of the lunar magma ocean had solidified. The residual liquid after the large degree of fractionation could not have maintained the chondritic REE pattern. Therefore the liquid from which the mare basalt source rocks precipitated must have had a LREE-enriched pattern. Our model does not require either significant amount of trapped liquid in the source rocks or an initial magma ocean that was unreasonably enriched in incompatible elements.

Although fractional crystallization and assimilation might have changed the mare basalt chemistries somewhat, the main reason for the diverse mare basalts types must be the different magma sources that were crystallized at various stages of the primary lunar differentiation.

It has often been assumed that about the outer 500 km of the moon melted about 4.6-4.4 b.y. ago [e.g., Tayler, 1978]. According to the differentiation process of the magma ocean shown in Figure 2, an ~20-km-thick anorthositic crust and ~150-km-thick dunite layer formed at the bottom of the magma ocean. At the top, some mafic minerals were incorporated with plagioclase and the other crust is thinner, and some mafic minerals were incorporated with plagioclase. In contrast, the high-Ti, low-K basalts have low Lu/Hf ratios and are enriched in Hf relative to Lu. The REE patterns of the low-Ti basalts are consistent with the thought that KREEP are residual liquids of the lunar magma ocean. The KREEPy highland basalts may represent the liquid of a highly evolved lunar magma ocean by various degrees of fractionation before assimilation processes or meteorite bombardments complicated their chemistries. The so-called uKREEP hypothesis is unnecessary. We emphasize that the VLT basalt and the green glass samples have very high Lu/Hf ratios, which shows that they were not derived from ‘primordial and chondritic’ material.

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