Petrogenesis of Lunar Rocks: Rb-Sr Constraints and Lack of H₂O¹

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Rb and Sr isotopic data and other chemical data indicate major lunar differentiation at about 4.6 AE (AE = 10° years) and very limited subsequent differentiation. The constraints of limited differentiation after 4.6 AE and the apparent lack of H₂O on the Moon, when applied to the derivation and petrogenesis of lunar samples, suggest the following: (1) soil samples, breccias, metaclastic rocks, and feldspathic basalts represent mixtures of repeatedly modified clastic material, which was ultimately derived from materials formed during the \sim 4.6 AE differentiation; and (2) mare basalts crystallized from melts which formed by partial melting, and which developed without equilibration between the melt and crystalline residuum.

Rb-Sr mineral isochrons currently provide the basic chronology of lunar evolution (refs. 1-14). Rb/Sr data also impose rigorous constraints on lunar petrogenetic models. This paper will discuss these constraints, emphasize the important role of large-scale differentiation which occurred at about 4.6 AE, and show that only limited chemical fractionation occurred during the subsequent evolution of most lunar rocks. Regardless of other types of evidence, no petrogenetic theory for the origin of lunar rocks can invoke extensive fractionation later than about 4.6 AE as a dominant part of the theory. The lack of H₂O on the Moon may be the critical physical-chemical factor limiting subsequent fractionation in many processes.

Rb-Sr Systematics and Fractionation Factors

Measurements of the isotopic abundance of Rb and Sr in the various mineral phases of a rock provide information not only on the time of crystallization and equilibration (T_x) , but also on the fractionation history of the rock prior to this most recent crystallization and equilibration. As illustrated in figure 1, cogenetic systems, either consanguineous total rocks or the various minerals in a single rock, attain identical values of 87 Sr/ 86 Sr, but during equilibration at T_x , different values of 87Rb/86Sr. On the Rb-Sr evolution diagram these different compositions subsequently evolve along straight line trajectories with a slope of -1. If the systems were closed to gain or loss of Rb and Sr since T_x , then the cognetic systems measured at any time form a linear array on the Rb-Sr evolution diagram. An array based on minerals from a single rock is a mineral or internal isochron, and one based on cogenetic rocks is a total-rock isochron. The isochron has a slope indicative of the time since equilibration (slope = exp (λT_x-1)) and an 87Sr/86Sr intercept, (87Sr/86Sr), equal to the Sr isotopic composition at time T_x (ref. 15).

The deviation of $(^{87}Sr)^{86}Sr)_1$ from that assumed to have existed at some time prior to T_x , coupled with the $^{87}Rb/^{86}Sr$, provides

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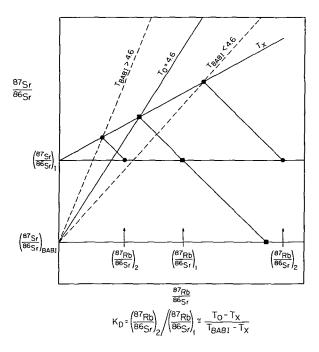


Figure 1.—Rb-Sr evolution diagram. Material formed at T_o with (**Sr/**sr) equal to (**Sr/**sr)_{BABI} is represented by a square. Fractionation at time T_x results in a portion enriched and a portion depleted in Rb relative to Sr (circles) and an unfractionated portion (square), all of which lie along the T_x isochron. The unfractionated portion yields a model age, T_{BABI} , equal to T_o , whereas fractionated portions yield model ages different from T_o .

an integrated measure of the Rb/Sr fractionation history of the rock. This fractionation history can be parameterized by a twostage model as illustrated in figure 1. The model assumes that a source material originated at reference time $T_o = 4.6$ AE with the "BABI" value of 87Sr/86Sr ((87Sr/ 86 Sr)_{BABI} = 0.69898) (ref 16). Fractionation at time T_x resulted in three fractions, one enriched in Rb relative to Sr, one unfractionated, and one depleted in Rb relative to Sr. Mineral isochrons on all three rocks would yield identical ages (T_x) and the same (87Sr/86Sr)₁. However, they would have different model ages, T_{BABI} , which is the time required for the 87Sr/86Sr of the total rock with its measured 87Rb/86Sr to evolve from $(87Sr/86Sr)_{BABI}$. The unfractionated rock will have $T_{BABI} = 4.6$ AE, the enriched rock will have $T_{BABI} < 4.6$ AE, and the depleted rock will have $T_{BABI} > 4.6$ AE. Thus, any deviation of T_{BABI} from 4.6 AE indicates a fractionation history prior to T_x .

The fractionation factor for this two-stage model is (ref. 10)

$$K_D \equiv \frac{(87 \text{Rb}/86 \text{Sr})_2}{(87 \text{Rb}/86 \text{Sr})_1} \cong \frac{T_o - T_x}{T_{BABI} - T_x}$$

This approximation is quite accurate since the decay constant for Rb is small. During the time interval from T_o to T_x numerous episodes of fractionation could have affected the rock as opposed to the simple two-stage model illustrated. However, ($^{87}\text{Rb}/^{86}\text{Sr}$)₁ is still the integrated $^{87}\text{Rb}/^{86}\text{Sr}$ in the interval from T_o to T_x .

As noted previously $T_o = 4.6$ AE and $(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_{T_o} = 0.69898$ are reference values, and the subsequent conclusions drawn in this paper are basically independent of their precise value. In fact, the time of major differentiation is probably not 4.6 AE, but may be as low as 4.5 AE or even 4.4 AE (ref. 13).

Lunar Rock Groups

On the basis of petrologic characteristics seven different groups of lunar rocks are recognized. Each of these groups has a distinctive Rb-Sr isotopic pattern. The Rb-Sr data are summarized in figure 2, which shows T_x , T_{BABI} , and K_D for representative members of each group. Most of the type examples shown are those on which we have made detailed petrographic and electron probe studies in conjunction with the Rb-Sr isotopic studies of Papanastassiou and Wasserburg. Figure 2 indicates that six of these groups are characterized by T_{BABI} close to 4.6 and $K_D < 2$. The seven groups are as follows:

1. Soils with $T_{BABI} = 4.6 \pm 0.3$ AE

Clots from the soil samples and friable soil-breccia samples, as well as bulk soil samples, have model ages of about 4.6 AE. This group includes samples from all landing sites and has a wide range of Rb/Sr (ref. 10). To a large extent many of these model ages

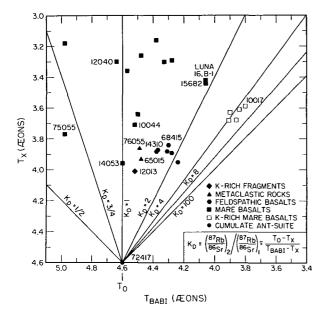


Figure 2.— $T_{\rm BABI}$ versus $T_{\rm x}$. Despite the variety of rock types represented, nearly all samples indicate less than a factor of 2 fractionation of Rb relative to Sr subsequent to $T_{\rm o}=4.6$ AE. Only the K-rich mare basalts indicate a greater degree of fractionation.

are dominated by a small fraction of very high Rb/Sr material with a model age of about 4.6 AE (ref. 10).

2. K-rich fragments with $T_{BABI} = 4.3$ to 4.6 AE

These fragments, the so-called "KREEP" rocks (ref. 17), include glass-rich agglutinates and metaclastic rocks, and have been found in the soils at all landing sites. Most are small fragments such as Lunar Rock 1 (ref. 18), and no internal isochrons have been measured on them. Nyquist et al. (ref. 19), however, showed that by grouping such fragments by chemical composition and location, Rb and Sr data yield linear arrays, which, if interpreted as total-rock isochrons, indicate ages ranging from 4.1 to 4.4 AE. Sample 12013 is the only large sample which we would place in this group. It is a heterogeneous metaclastic rock with K, Th, and U concentration a factor of 40 greater than typical mare basalts and a factor of 10 greater than Apollo 11 K-rich basalts (ref. 20). Fragments of 12013 have a model age

of 4.52 AE and a recrystallization age (T_x) of 4.01 AE (ref. 2).

3. Metaclastic rocks with $T_x \approx 3.95$ AE and $T_{BABI} \approx 4.5$ AE

This group includes a large proportion of the Lunar Highlands samples and also constitutes a large proportion of the lithic fragments in soil samples from all landing sites. These clastic rocks, composed predominantly of plagioclase, have been extensively recrystallized by metamorphic and/or partial melting processes (ref. 21). Typical examples are 65015 and 76055, both of which display isotopic and petrologic evidence for extensive, but not complete, equilibration at 3.95 AE (refs. 10, 13, 21, and 22). Step-wise heating ⁴⁰Ar-³⁹Ar studies on 65015 suggest that the cores of the larger plagioclase clasts have an age greater than 4.46 AE (ref. 22). This is also suggested by Rb-Sr isotopic data (ref. 10).

4. Feldspathic basalts with $T_x \approx 3.85$ AE and $T_{BABI} \approx 4.3$ AE

This group includes a number of samples of intersertal, plagioclase-rich basalts from the Apollo 14 and 16 landing sites (e.g., 14310, 14276, and 68415). In addition to the high plagioclase content (60 to 80 percent) they are characterized by high K, rare earth element, P, Ba, U, and Th contents (ref. 23), and a high content of siderophile elements (ref. 24). Even in these rocks, which almost certainly crystallized from a melt, plagioclase grains are present which, on the basis of electron probe data, have not completely equilibrated with the melt (ref. 23). 40Ar-³⁹Ar studies also indicate older relict plagioclase and provide evidence for an older event (refs. 25 and 26).

5. Mare basalts with $T_x = 3.16$ to 3.95 AE and $T_{BABI} = 4.1$ to 5.0 AE

This group includes all of the mare basalts with the exception of those in Group 6. Samples from each landing site have similar T_x and T_{BABI} , but $(^{87}Sr)^{86}Sr)_1$ values and trace element concentrations differ for samples from an individual landing site (refs. 6, 11, 12, 13, and 27). This suggests derivation of individual samples (and flows) from different sources (ref. 27) or differing degrees

of assimilation of country rock (ref. 6). Typical well-characterized samples from the various landing sites include:

10044 (refs. 1, 28, and 29) 12040 (refs. 6, 30, and 31) 14053 (refs. 7, 32, and 33) 15682 (refs. 11 and 34) 75055 (refs. 13, 21, and 26) Luna 16, B-1 (refs. 8, 35, and 36).

6. Mare basalts with $T_x = 3.65$ AE and $T_{BABI} = 3.85$ AE

Although grossly similar to the Apollo 11 low-K basalts included in Group 5, these samples from the Apollo 11 landing site are higher in K and other incompatible elements, and have much younger model ages. A typical well-characterized example is 10017 (refs. 1, 29, and 37).

7. "ANT" rocks with $T_{BABI} = 4.6 \text{ AE}$

"The "ANT" rock suite includes the coarse-grained rocks of the anorthositenorite-troctolite-dunite suite. In general they display magmatic cumulate textures, but are extensively modified by shock processes. Dunite sample 72417 has both an isochron age and a model age of about 4.6 AE (ref. 3). No mineral isochron ages have been measured on anorthosite samples such as 15415 (refs. 38 and 39), or on troctolite samples such as 76535 (ref. 40). However, low 87Sr/86Sr ratios indicate that these rocks cannot have equilibrated and resided in a higher Rb/Sr environment for any extended length of time (ref. 14; and Papanastassiou, personal communication).

Nature of Major Differentiation at ~ 4.6 AE

Many types of chemical, isotopic, and physical evidence are consistent with the hypothesis of primitive, large-scale, crustal differentiation (refs. 1, 41, 42, and 43). The presence of a "magic component" (ref. 5), with T_{BABI} of 4.6 AE and a high ⁸⁷Rb/ ⁸⁶Sr which dominates the model age of many lunar soil samples, indicates that this differentiation occurred at about 4.6 AE and produced rocks with very high Rb/Sr ratios.

The existence of high Rb/Sr material with relatively old model ages was confirmed by the discovery of the K-rich rock 12013 (ref. 2) and other fragments (refs. 17 and 18). The existence of material complementary to the Rb/Sr-rich material is indicated by dunite sample 72417, which has crystallization and model ages of 4.6 AE (ref. 3).

Rb-Sr data also indicate that only limited fractionation occurred, subsequent to the primitive differentiation, and furthermore suggest that most of the observed chemical characteristics were produced during the primitive differentiation. That most of the chemical differences observed in the lunar rocks are consistent with primitive differentiation at ~ 4.6 AE, rather than subsequent fractionation processes, is illustrated in figure 3. Sm/Eu, a parameter sensitive to frac-

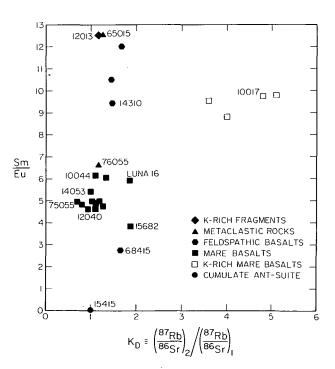


Figure 3.—Sm/Eu versus K_D. The large range of Sm/Eu, indicative of extensive fractionation, is not commensurate with the fractionation of Rb relative to Sr as indicated by the small range of K_D. This indicates that the fractionation of Sm and Eu occurred prior to T_x; and, from additional data, most likely occurred during the large-scale lunar differentiation at ~4.6 AE. Data on Sm and Eu are from the following: refs. 17 and 44-58.

tionation, varies by a factor of ~ 200 , whereas K_{p_1} a measure of fractionation after the differentiation at ~ 4.6 AE, varies only by a factor of about 2. The strong fractionation indicated by Sm/Eu must have occurred prior to the time of crystallization. Although neither K_D nor Sm/Eu is particularly sensitive to olivine or Ca-poor pyroxene fractionation, both are extremely sensitive to fractionation of plagioclase or of late-stage K-rich material. The large range of Rb/Sr observed between samples, approximately a factor of 1000, is comparable to the range of Sm/Eu. Hence, we conclude that the Sm/Eu differences must have been a characteristic of the source from which the rocks were derived and that most chemical differences in lunar rocks are a result of primitive differentation at about 4.6 AE.

Regardless of physical details, this primitive differentiation process resulted in a fraction rich in K (Rb. Ba. U. Th. trivalent rare earth elements), a Ca-Al-Si-rich fraction (anorthosite and anorthositic gabbro), and a Mg-Fe-rich fraction (dunite, troctolite, and norite). Samples of the Ca-Al-Si-rich fraction and the Mg-Fe-rich fraction have survived subsequent excavation by meteorite impact, but exhibit a wide range of modification. Less modified samples suggest that these fractions cooled slowly enough to produce rocks with coarse-grained, homogeneous phases. The original nature of the K-rich fraction is not clear as no samples have been recognized which have not been extensively modified.

Possible Petrogenetic Processes Involving Limited Fractionation

The Rb-Sr constraint on the amount of fractionation, as well as constraints imposed by many other kinds of data, are satisfied if we hypothesize that soils, glass-agglutinate fragments, friable breccias, and progenitors of metaclastic rocks and feldspathic basalts (Groups 1–4) are all basically mixtures of clastic material, which have been subsequently modified by a variety of processes,

including fragmentation. metamorphism. partial melting, and complete melting. Rb and Sr would not be fractionated if the formation of the clastic mixture involved only fragmentation of preexistent rocks from one or many sources, even if fragmentation occurred repeatedly over a long period of time. Consequently, if the source regions of a clastic mixture are primary, unmodified materials formed during the primitive differentiation at ~ 4.6 AE, or if they themselves are clastic mixtures repeatedly modified either by continued fragmentation and mixing, or by processes characterized below, then Rb and Sr will remain unfractionated and the model age of the mixture will still reflect the time of primitive differentiation.

Preservation of the old model age of such a mixture would be accomplished during subsequent modification by processes with the following characteristics, even if repeated many times:

- 1. Volatile loss of Rb was in general not significant.
- 2. Metamorphism, in the absence of H_2O , was strictly controlled by solid-state and grain-surface diffusion and resulted in lithification by sintering at grain boundaries with only short-range migration and limited segregation of elements.
- 3. Partial melting in the metaclastic rocks of Group 3, and in the K-rich fragments of Group 2, was characterized by extensive reaction between an intersitial melt and larger clastic grains. Lack of Rb-Sr fractionation dictates limited mobility of the melt and only short-range migration of elements within the melt. These characteristics can be partially attributed to the absence of H₂O and to the fine-scale homogeneity of the fragmental mixture. Local segregation of Rb-rich material and partial Sr equilibration are suggested by the Rb/Sr data of Nyquist et al. (ref. 19) on small chemically defined groups of samples from single sites. The Rb/Sr data on these sam-

- ples have been interpreted as total-rock isochrons representing distinct events at times ranging from 4.1 AE to 4.4 AE. These may alternatively be interpreted as the result of local segregation of K-rich, Rb-rich material without total Sr equilibration at 3.95 AE.
- 4. Impact-produced melts, which formed by nearly total melting of soil, breccia, or metaclastic rocks, crystallized as the feldspathic basalts of Group 4. Such an origin would preserve the old model age of the source and would satisfy several other geochemical constraints on these rocks, such as the high content of siderophile elements. However, the Rb-Sr constraint could also be satisfied if these rocks formed by partial melting of plagioclase-rich source rocks with the additional restrictions described below for mare basalts.

The Rb-Sr constraint limiting fractionation is satisfied if the ultimate source of the clastic mixture formed during the large-scale differentiation at ~ 4.6 AE. If the modification process or processes retain the characteristics described above, or if modification is a simple fragmentation process, then fractionation does not basically occur and old model ages are preserved. This is true regardless of either the order or the number of times this clastic mixture is modified.

Mare basalts with near 4.6 AE model ages (Group 5) must also have been derived without substantial fractionation of Rb and Sr, either during formation of the parent magma or during the ascent and crystallization. All other chemical parameters suggestive of a greater degree of fractionation must be a characteristic of the source region. The origin of the mare basalts is further restricted by the (87Sr/86Sr)₁ values, which suggest that rocks of the same age were derived from a number of different sources. A magma meeting these requirements could be produced by several mechanisms (ref. 23):

1. Total melting of a source rock which has an Rb-Sr model age of 4.6 AE and also meets all other chemical and iso-

- topic constraints would form a rock satisfying the Rb-Sr constraints. Although total melting is generally regarded as an unlikely terrestrial event, it is possible that, in the absence of $\rm H_2O$ and tectonic activity, instability and separation of a melt from a source region would be delayed until complete melting occurs. *Total* melting could also occur as a result of impact processes.
- 2. Uniform contamination of relatively low Rb/Sr melts by assimilation of Rbrich crustal material with a model age of 4.6 AE is the mechanism invoked by Papanastassiou and Wasserburg (ref. 6).
- 3. Our preferred hypothesis is that, in the absence of H₂O, partial melting occurs by incremental melting of integral volumes of solid phases with little or no equilibration between melt and crystalline residuum. Thus, the low-temperature phases rich in Rb and 87Sr would melt totally and grains of higher temperature phases would melt peripherally, but the solid residuum would not equilibrate with the melt. As pointed out by Graham and Ringwood (ref. 59), the resulting melt would have the same model age as the source region. Any crystallization and separation of Ca-rich pyroxene and/or plagioclase during the ascent of the melt to the surface would result in Rb-Sr fractionation. However, silicate melt curves typically have a positive slope (ΔP) $\Delta T > 0$) in the absence of H₂O, and under these circumstances the melt may become superheated as it moves upward, effectively preventing crystallization and consequent Rb-Sr fractionation. Production of a superheated magma is also an important consideration in the contamination hypothesis. since it would facilitate assimilation and homogenization.

The Apollo 11 K-rich mare basalts (Group 6) could also form by this process, but the younger model ages require a greater degree

of equilibration between the melt and residuum or of some fractional crystallization before extrusion onto the surface.

Conclusion

An intriguing feature of these explanations for deriving lunar rocks without fractionation of Rb and Sr is the linking of this special characteristic to another characteristic lunar feature—the apparent lack of indigenous H_2O . The hypotheses outlined here differ from other models of lunar petrogenesis in that many rock types would be derived by near-surface modification of rocks formed during primitive crustal differentiation. This can be accomplished with energy derived partially from impacting bodies rather than totally from internal heat sources.

Acknowledgment

Most of the Rb-Sr patterns summarized here are based on mineral isochron data from G. J. Wasserburg and D. A. Papanastassiou. Furthermore, many of the arguments incorporated into these hypotheses have been expressed in their published papers and expressed even more vociferously in numerous discussions during the course of our cooperative work since July 1969. Unpublished Sm and Eu data were provided by N. J. Hubbard and by J. A. Philpotts. This paper has been supported by NASA grants NGL-05-002-188 and NGL-05-002-338.

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