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Lunar evolution: is there a global radioactive crust on the Moon?

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Chemical and isotopic analyses of various grainsize fractions of lunar soils show the presence of an 'exotic component' in practically all lunar soils. The patterns of enrichments in the grain-size fractions and the Sr-isotopic data show that the regolith evolution displays the combined effects comminution of local rock types and addition of the exotic component. The chemical characteristics of this exotic component as deduced from the chemical and isotopic data in soils from Apollo 11, 12, 15 and 16 uniformly point to compositions similar to the material from Fra Mauro region collected in the Apollo 14 mission. There is a strong correlation between the amount of exotic component in a soil and its distance from the Fra Mauro region. It is suggested that the exotic component represents trace element enriched material from the Imbrium-Procellarum region, which was surficially deposited during Imbrium excavation and re-exposed from under the mare-lavas in subsequent cratering events. Surficial transport processes have distributed these materials widely over the lunar surface. There appears no need to invoke a global radioactive crust on the Moon nor of 'hot spots' distributed over the entire surface of the Moon to explain the ubiquitous presence of this component in lunar regolith, nor is there a compelling reason at present to postulate a global melting process for the generation of highly differentiated materials such as 'kreep' and the exotic component.

Introduction

The concept of a radioactive crust on the Moon was first introduced by Papanastassiou & Wasserburg (1970) based upon their discovery that the lunar soil at Tranquillity Base contained a 'magic component' characterized by enrichments in some lithophile trace elements. It is now clear that practically all lunar soils show the presence of such a component, in variable proportions relative to local rock debris. These observations thus called for the occurrence of highly differentiated materials on the Moon easily available for widespread transport over the lunar surface. We call this the 'exotic component' primarily in the sense that in most lunar regolith this component appears to be of non-local provenance. It is to be noted that actual rock fragments closely resembling the exotic component have been discovered in the soils. Thus, Schnetzler, Philpotts & Bottino (1970) and Hubbard, Gast & Wiesmann (1970) have suggested that the dark portions of rock 12013 are pure exotic component. In the Apollo-12 soils, Hubbard, Meyer, Gast & Wiesmann (1971) discovered small rock fragments which they termed 'kreep' in view of the high abundances of potassium, rare earth elements and phosphorous in these rock fragments.

A fundamental question that arises is whether these highly differentiated materials, sampled as the exotic component in lunar fines and as discrete rock fragments represent a moon-wide radioactive crust, or whether their source is more regional than global. The former possibility has been extensively dealt with by Papanastassiou & Wasserburg (1970) to postulate the currently prevalent ideas of global lunar differentiation at or soon after accretion of the Moon at 4.6 Ga ago. I wish to explore here the characteristics of the exotic component in various lunar soils to discuss the origin and provenance of this differentiated materials in the context of lunar crustal evolution.

Characteristics of the exotic component in lunar fines

A characteristic of all lunar soils analysed by us (Murthy, Evensen, Jahn & Coscio, 1972; Evensen, Murthy & Coscio 1973, 1974) is that the finer grain-sizes of all soils contain relatively enhanced abundances of the trace elements K, Rb, Sr and Ba relative to coarser grain-sizes.

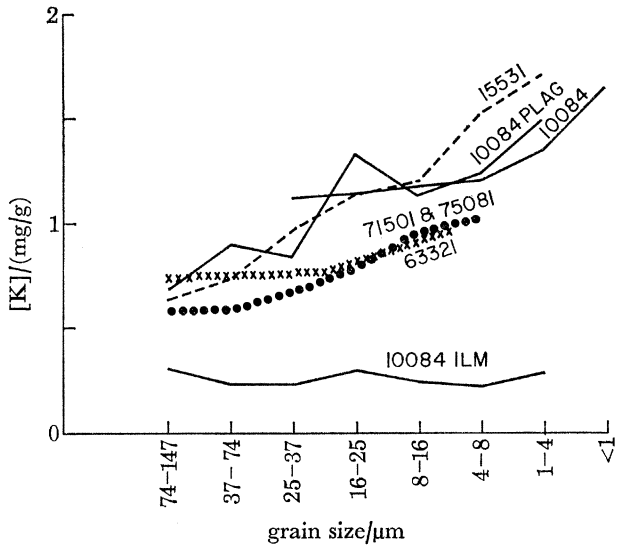

Figure 1. Concentration K in various grain size and mineral fractions of lunar fines from Apollo 11, 15, 16 and 17 sites.

In figure 1, a summarized version of our data of trace element abundances in the grain-size fractions of various lunar soils is shown. Only the K concentration versus grain size is given for the purpose of clarity, but entirely analogous situations exist for the other trace elements such as Rb, Sr and Ba (Murthy *et al.* 1972; Evensen *et al.* 1973, 1974). These enrichment trends can be produced by one or more of the following processes:

(1) Volatilization and gettering effects which preferentially trap these trace elements in the finer sizes.

(2) Differential comminution of local rocks with the release of trace element rich mesostasis and grain boundary films into the finer sizes.

(3) Preferential residence of the exotic component in the finer grain-sizes.

Volatilization and recondensation processes are shown to be important on the lunar surface by several workers (e.g. Naughton, Derby & Lewis 1971; Gibson & Hubbard 1972; Gibson *et al.* 1973; Chou, Baedecker, Bild & Wasson 1974; Clayton, Mayeda & Hurd 1974). We do not believe, however, that the presently observed enrichments can be ascribed to such processes, for the following reasons. First, not only the volatile elements but refractory trace elements such as Sr and Ba show the same trends as shown by volatile K and Rb. Secondly, in any volatilization process, fine grained material with a higher surface to volume ratio, should lose volatiles more efficiently than coarser grain-sizes, exactly the opposite of the trend observed here. Alternatively, gettering effects, which are surface area dependent, should show enrichments commensurate with the $1/r$ dependence of the surface to volume ratio of the various grain sizes. The actual enrichments observed are much less than any surface-correlated effects. Surface-correlated grain-size effects are also ruled out by the data of ilmenite from 10084 which shows no trends. These and other aspects of the Sr-isotopic data which is discussed later and the lack of K/Rb fractionation between size fractions as expected in volatilization processes (Gibson *et al.* 1973) indicate that these processes cannot explain the observed enrichment trends in lunar soils.

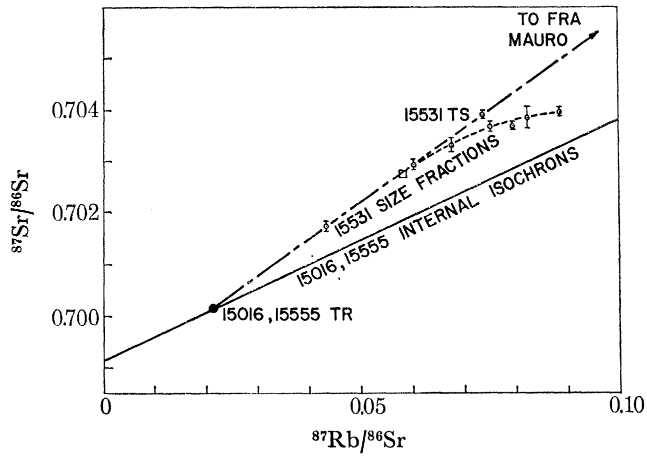

FIGURE 2. Rb-Sr isotopic systematics of grain-size fractions of lunar fines 15531. Grain size decrease from left to right. Point marked TS is measured on a total sample aliquot. Internal isochron line from Murthy *et al.* (1972) and Evensen *et al.* (1973).

We turn now to the second and third causes suggested above. It can be shown that comminution effects alone cannot explain all the observed features of the various grain-size fractions. The best evidence for this is provided by the Sr-isotopic characteristics of the grain-size fractions, as shown in figure 2 for the Apollo 15 sample 15531. The monotonic increase in K, Rb, Sr and Ba in progressively finer fractions is accompanied by an increase in Rb/Sr and $^{87}Sr/^{86}Sr$ ratios. If comminution were the sole process, the Sr-isotopic compositions of all the grain-size fractions should plot along a mixing line on the Rb-Sr evolution diagram which should coincide with the internal isochron of the local rocks such as 15555 and 15016. This, however, definitely is not the case.

By far the most reasonable explanation, therefore, is that an exotic component enriched in K, Rb, Sr and Ba has been added as well to the lunar soils. Thus, the isotopic and trace element characteristics, taken together, are attributed to a combination of comminution effects and mixing with the exotic component (Evensen *et al.* 1973). In figure 2, any mixture of the local regolith and the exotic component should describe a point on the mixing line connecting local rocks to the exotic component. Simple comminution would disperse the regolith along the internal isochron. The isotopic data however show that these two processes acting together can give rise to the nonlinear array of data points and show variable model ages bracketed by the age of the local rocks and the model age of the exotic component. The dispersion of the Sr-isotopic data in the coarser grain sizes along a 4.6 Ga reference line strongly suggests that the model age of the exotic component is about 4.6 Ga.

Since most of the lunar soils appear to be mixtures, a characterization of the pure end member exotic component is impossible. However, the finest grain-size fraction in each of the soils analysed by us can be used to set lower limits for the abundance of various trace-elements and more particularly, several diagnostic trace-element ratios of the exotic component. Some characteristics thus deduced are shown in table 1. In addition, the model ages of this component must be about 4.6 Ga, as noted above.

Table 1. Trace element ratios of finest grain-size fractions of lunar soils and comparison with Fra Mauro soil

sample site	size fraction µm	K/Sr	Rb/Sr	Ba/Sr
Apollo 17	4–8	5	0.01	0.75
Apollo 11	< 1	7	0.02	1.2
Apollo 15	1–4	11	0.03	1.4
Apollo 16	4–16	5	0.01	0.65
Apollo 12	—	18	0.06	3.5
Apollo 14	—	24	0.08–0.1	4.0–4.5

Source areas of the exotic component

While the general chemical and isotopic characteristics of the exotic component can be deduced from these studies, it is not, however, *a priori* evident how and where on the Moon such a component might be formed or how it is distributed so ubiquitously into soils.

The data from the lunar orbital γ-ray spectrometry reveals a definite association of high radioactivity areas in the Imbrium-Procellarum region (Metzger *et al.* 1973; also Plate II from the Proceedings of the *Fourth Lunar Science Conference*, vol. 1). In these areas, the highest activity is localized to several areas in the vicinity of Aristarchus, Archimedes and Fra Mauro. Of these locations, only Fra Mauro material was directly sampled at the Apollo 14 landing site.

It is of interest in this context to note a number of features of the Fra Mauro material returned by Apollo-14 mission. The materials sampled here have highly radiogenic $^{87}Sr/^{86}Sr$ ratios and model ages close to 4.6 Ga. The trace element ratios and the elemental abundances are such that they can clearly match the characteristics of the inferred exotic component in the lunar soils (table 1). If the Fra Mauro formation as a whole is similar to the materials sampled in Apollo 14 mission, it is clear then that here is a surficial exposure of a large amount of material, available for transport into the other regions by various lunar surficial processes. We have therefore suggested earlier (Murthy *et al.* 1972, 1973) that the Fra Mauro formation could be the source of the exotic component observed in lunar soils.

It appears, tentatively at least, that the Fra Mauro and perhaps the other areas of high radioactivity identified in the orbital γ spectrometry experiments, could serve as the source of the exotic component observed in varying amounts in lunar soil samples. These surficially exposed materials, if transported over wide areas of the Moon and mixed with locally derived regolith, can as a first approximation explain all our chemical and isotopic data on the lunar soils. It seems inevitable that such transport must have occurred to some degree. Certainly fragments of presumably highland derived anorthositic fragments have been transported to mare sites as first noted by Wood, Dickey, Martin & Powell (1970). For the extremely fine-grained material, the transport must have been more efficient. For example, from the K-Ar studies on mineral separates from Apollo-11 soil 10084, Basford (1974) notes that significant amounts of fine grained material may have been transported over distances of the order of several hundreds of kilometres.

A test of the adequacy of the above hypothesis which postulates that the source area for the exotic component is the region of the high radioactivity areas in the Imbrium-Procellarum region, is to seek a correlation between the abundance of exotic component and distance from

TABLE 2

	% exotic component†	
	1	2
Apollo 14	100‡	100‡
Apollo 11	12	13
Apollo 12	37	59
Apollo 15	14	37
Apollo 16	9–18§	9–18§

† % = [Rb (soil)–Rb (rocks)]/[Rb(A-14 soil)–Rb (rocks)].
‡ Assumed to be the pure exotic component.
§ Rock values are highly dependent upon proportions of different rocks assumed in this geologically complex site. Also, the regolith at this site is 2 times thicker than at other sites. Percentage exotic component is dependent on mixing depth in local regolith. Higher value corresponds to complete mixing with full depth of regolith.
Column 1 is from Evensen *et al.* (1974). Column 2 uses rock and soil values compiled by Rose *et al.* (1974).

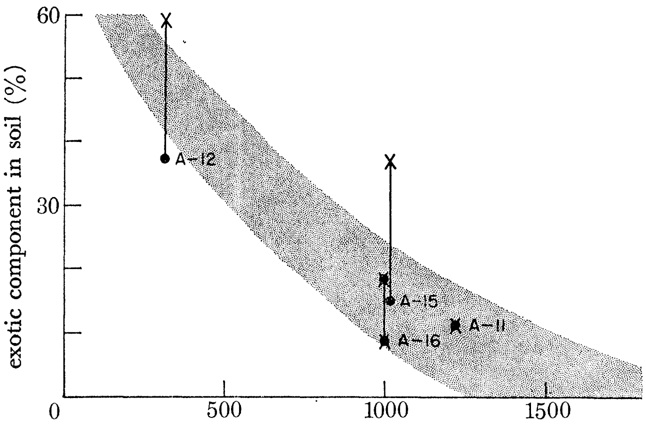

FIGURE 3. Percentage of Fra Mauro type exotic component in soils from Apollo missions plotted against distance from Apollo 14 landing site at Fra Mauro. Percentages of exotic component and uncertainties in the estimates as discussed in the text and in Evensen *et al.* (1974). ●, Evensen *et al.* (1974). ×, average rock and soil values from Rose *et al.* (1974).

the source. Unfortunately, only material from Fra Mauro has been sampled, and the chemical and isotopic characteristics of material from the other radioactive areas in the region are not directly known. Assuming, however, that all these materials are similar to Fra Mauro, and that local rocks of each site represent the *in situ* material, we can roughly estimate the amount of exotic component at each site. Obviously, a simple two component model of this type can only be a crude approximation to the complex mixing processes in the lunar regolith.

Using Rb as a diagnostic element, we have estimated the percentage of exotic component in various lunar fines (Evensen *et al.* 1974). It is realized of course, that the estimation of average abundances of an element in rocks and soils is somewhat difficult, particularly in complex regions such as the Apollo-16 site. In the present estimate (table 2) the averages of rocks and soils measured in our laboratory (Evensen *et al.* 1974) have been used to minimize the problem of local variability of rocks and soils, but we emphasize that the generality of relationship proposed here remains, although the absolute percentages of the exotic components may vary, depending upon the choices used for averages. We also note that in spite of the vagaries of the simplistic two-component model used here, the estimates obtained are in reasonable agreement with those deduced from much more refined multicomponent models (e.g. Schoenfeld & Meyer 1972; Wänke *et al.* 1972; Wasson & Baedecker 1972). In figure 3, these estimated percentages are plotted against distance from Fra Mauro area. The inverse correlation between distance and the percentage of exotic component seems remarkable, especially that neither the source area nor the estimated percentages are precisely definable parameters. Of course, the actual source area is not confined to Fra Mauro; the dominant contribution at Apollo-15 site, for example is from the Archimedes area.

Relation between the exotic component and kreep

Petrologic and chemical studies of Apollo-11 and 12 soils have shown the existence of lithic fragments and glasses having high trace-element abundances (e.g. Albee & Chodos 1970; Hubbard *et al.* 1971; Keil, Prinz & Bunch 1971; Wood *et al.* 1971). These materials are identical to basaltic glasses in the Fra Mauro soils (Apollo Soil Survey 1971). These chemical and more recent isotopic analyses (Nyquist, Bansal & Weismann 1975) show that various kreep materials recognized in these sites are similar to each other, and match well the characteristics of the fine grained exotic component in the soils. While retaining the distinction that kreep materials are petrologically defined fragments and that the exotic component is material inferred from trace-element analyses of soils, we can examine the relationship between the exotic component and kreep. Fra Mauro materials are considered to be essentially pure kreep (Hubbard *et al.* 1972). The isotopic systematics of the soil fractions reported here indicate its presence as a component in the soils. Furthermore, the petrologic and chemical affinities of kreep to feldspathic compositions has been well documented by numerous investigators. This seems also true for the exotic component, as our data on mineral separates from Apollo 11 soil fractions show a close match between the trends exhibited by the bulk soil and the plagioclase size fractions (figure 1). Thus, we can consider the exotic component as fine grained kreep. Dissemination of fine grained exotic component into lunar soils must be accompanied by the dissemination of lesser amounts of coarse sized kreep fragments as well.

Relationship of exotic component and kreep to Imbrium basin

The materials at Fra Mauro and the other high γ ray activity areas in the Imbrium-Procellarum area are considered to be derived from the Imbrium basin excavation (Sutton, Hart & Swann 1972). Since Imbrium is the largest and deepest of the circular mare basins on the Moon, it is likely that its excavation has brought up material from deeper in the lunar interior or under more thermally energetic situation than in other events. Such excavated material would initially be found as ejecta surrounding the Imbrium basin, but over the last 4 Ga would diffuse outward into the surrounding regolith at a rate controlled by the nature of lunar transport processes. Such processes for example, are reasonably effective at transporting anorthosite and other lithologies into mare regions, and presumably transport fine grained material more efficiently. The primary Imbrium ejecta would have been buried under the younger mare filling in other basins. Such buried material can be tapped for redistribution, perhaps in diluted form, only in cratering events that punctured through the mare filling. Examples of this are the Archimedes and the much later Copernicus craters. Through such primary and later sources, distribution of the exotic component ultimately results in a broad pattern centred around the Imbrium region as clearly identified in the orbital γ ray spectrometry experiments (Metzger *et al.* 1973).

In view of the chemical and isotopic similarities between kreep and the exotic component, the general correlation between abundance of exotic component and the proportion of identifiable kreep fragments in the soils and the feldspathic affinities of both kreep and the exotic component, it is tempting to speculate that kreep distribution has a general history similar to that for the exotic component, particularly since no evidence for localized reservoirs other than Fra Mauro material has been found. The lunar soils can then be pictured as locally derived material dominated by the mare basalt-highland anorthosite bimodal tendency, overprinted with distance dependent kreep-exotic component, with additional vertical and lateral mixing provided by lunar gardening and transport processes.

If the above model is correct, it implies that either the kreep and exotic component material was present at a depth such that it was sampled only by the excavation of the Imbrium basin or that it was produced primarily as a consequence of the Imbrium impact event. If present at depth, it is not clear though that such a layer need be uniformly present over the lunar globe. It may be localized for example to the earth facing side or even to the Imbrium basin region. Lateral inhomogeneity seems to be the rule rather than the exception in the case of the Moon, as for example in the mare distribution, centre of mass and the principal moments of inertia.

The origin of the kreep-rich materials has been an enigma in lunar petrology and geochemistry. Based on the excellent work of Walker, Longhi & Hays (1972) and Walker *et al.* (1973), it is clear that the major element compositions of these materials can be accounted for by partial melting of rocks consisting of plagioclase, olivine, and low Ca-pyroxene, i.e. rocks similar to lunar anorthositic gabbros. If the lunar crust had already formed by segregation from a molten Moon, what was the energy source to remelt these anorthositic material to produce kreep by partial melting? Wood (1974) suggested that anorthositic crustal materials were subducted into the lunar mantle by global convective systems early in the history of the Moon, when the lunar crust was thin and weak. Kreep-rich material then would be produced in a process analogous to terrestrial andesitic volcanism. If it is such a generalized process, we would expect to observe a more generalized occurrence of kreep-rich materials on the lunar surface, and their particular association with Imbrium area is not explained. We suggest that the heat source needed was the impact energy of the colliding planetesimals and that in general, partial melting processes involved in production of exotic component and kreep-rich materials are local processes initiated by deposition of thermal energy by incoming planetesimals (Murthy & Banerjee 1973) into a solid lunar crust. The role of accessory phases and grain boundary films which carry substantial amounts of large-ion lithophile elements is significant in this context, whether these phases were produced in the moon because the prinicipal lunar mineral phases do not readily accommodate these elements (Taylor & Jakes 1974) or whether these phases were integral constituents of accreted lunar material. A transient and local heating such as in a collision event could induce complete melting of these phases to produce liquids which are rich in trace elements, without equilibrating with the solid residue in the source regions, in a manner analogous to that first described by Graham & Ringwood (1971). In the case of an event of large magnitude, the thermal energy may be sufficient to induce partial melting of the bulk solid consisting of plagioclase orthopyroxene and olivine. A mixture of the total melts of accessory phases and partial melts of the major phases would ensue. Alternatively, I suggest that the accessory phase melts produced can themselves induce partial melting of the bulk solids to some extent. The observational data on partial melts produced on grain surfaces by glass splatters (Grieve & Plant 1973) certainly strengthens this suggestion! Further, as pointed out by Albee & Gancarz (1974) anhydrous melts may become superheated as they move upward. Thus, the accessory phase total melts produced from some depth and their rapid segregation upwards, can lead to a situation where the superheated melt of accessory phases can cause grain surface melts of major silicate phases. The range in major element compositions represented by kreep compositions on the pseudo-ternary silica-anorthite-olivine diagram of Walker *et al.* (1973), the inverse correlation between Al_2O_3 and Eu and trivalent rare earths (Hubbard *et al.* 1974) and the wide range in MgO/FeO ratios (Walker *et al.* 1973) can be accommodated in this model by variations in the degree of dilution of accessory phase melts by the melts produced from major-minerals of the source region.

A corollary and consequence of the present model of kreep generation would be a variability of both ages and initial $^{87}Sr/^{86}Sr$ ratios of kreep materials, as pointed out by us earlier (Murthy & Banerjee 1973). Presently available isotopic data are consistent with this view (Nyquist *et al.* 1973, 1975). On these considerations, it appears that the exotic component observed in lunar soils in Apollo 11, 12, 14, 15 and 16 belongs to one parent material (or a series of related precursors), possibly confined to the Imbrium-Procellarum regions and dispersed widely on the lunar surface during Imbrium collision and subsequent cratering events such as Archimedes and Copernicus. The Apollo-17 kreep which has been sampled as noritic breccias from the north and south massifs appear to be ejecta from the Serenitatis basin. The exotic component in A-17 soils must surely be related to this local rock type, in addition to material transported from the Imbrium-Procellarum regions. McGetchin, Settle & Head (1973) have shown that the Imbrium ejecta is pervasive in all the Apollo landing sites. Considerable support for this suggestion also exists in the study of Morgan *et al.* (1974) which showed on the basis of the siderophile element distribution, that Imbrium ejecta are the dominant layer occupying the top most stratigraphic unit at Apollo-17 site. It is conceivable that the Apollo-17 kreep also may in part be from Imbrium region, although this is an open question at present.

The general distribution of kreep and the exotic component in the soils can thus be from a single source region in the Imbrium-Procellarum area. Imbrium excavation has widely dispersed these materials on the near side surface of the Moon. The material so dispersed has been reworked in highland sites in subsequent cratering events and dispersed even more effectively. Mare flooding has covered these ejecta in several basins, but the trace element-rich material has been re-excavated from beneath the lava flows occasionally as for example in Aristarchus.

If the distribution of highly differentiated material, such as kreep and the exotic component in fact have a single provenance (Imbrium-Procellarum region) as postulated here, the question remains whether the Imbrium excavation tapped a global layer at some depth or simply a local heterogeneity of composition. Nyquist *et al.* (1975) have convincingly argued that all the kreep materials indicate a single age of differentiation at ~ 4.25 Ga ago, and the present arguments call for a single provenance for all these materials. In this context, the Gargantuan Basin hypothesis is very appealing, because it allows for a pre-Imbrium lava flow caused by an early basin forming impact. Such a surficial layer at the Imbrium-Procellarum region can be excavated, impact-melted and distributed as ejecta at the time of Imbrium collision. Presently known isotopic and trace-element data are consistent with this point of view. There seems no compelling reason at present to postulate a global melting process for the generation of these highly differentiated materials on the Moon.

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