

cated that it is made up primarily of fine-grained basalt, glassy impactite material, and some crystal fragments. Samples 86,A and 86,B may contain different proportions of coarse and fine fragments and, hence, have different total carbon abundances. The fine-grained breccia appears to consist primarily of compacted fines, and it apparently lost no carbon during lithification.

The rare gas analyses reported in the preliminary examination of lunar samples from Apollo 11 (2) also showed enrichment in the fines and breccia. Assuming that all the rare gas content is due to solar wind, some rough calculations may be made to indicate how much of the carbon may be attributed to solar wind. The major unknown in the calculation is a "sticking" or retention factor relating the relative retention on the lunar fines to each of the rare gases. A comparison of Apollo 11 rare gas abundances with Cameron's 1967 (3) table of abundances in the solar system indicates a fractionation in the lunar materials favoring the retention of higher atomic weight gases. If the solar carbon/neon is normalized to the neon in the lunar samples, the solar wind contribution of carbon to the lunar fines is 6 $\mu\text{g/g}$ or about 4 percent of the total carbon. If the carbon abundance is calculated from the krypton abundance, the solar wind contribution is about 50 $\mu\text{g/g}$, and, if it is calculated from xenon, the solar wind contribution would be 300 $\mu\text{g/g}$. Assuming a similar "sticking" factor, the solar wind contribution would be about 5 times lower for nitrogen than for carbon.

A general calculation may also be made to estimate the possible meteoritic and cometary contribution to the lunar fines. The major unknown in this case is the composition of the meteoritic influx. A useful start might be to take material with the composition of carbonaceous chondrites. Such a choice is strengthened by comparing the increase in nickel from the lunar igneous rocks with the lunar breccia and fines as reported in the preliminary examination of samples from Apollo 11 (2). Nickel and carbon both show significant increases, and nickel, like carbon, has a much higher concentration in carbonaceous chondrites than in lunar rocks.

Mason's (4) review of carbonaceous chondrites gives typical concentrations of 2 percent carbon and 1 percent nickel in these meteorites. Using these values to calculate the approximate

contribution of meteoritic extra-lunar material indicates that about 0.5 to 2 percent has been mixed with lunar material. The nitrogen contribution of meteorites is about 10 percent that of carbon and, again, this element does not give an apparent satisfactory material balance. Based on our simplifying assumptions, both solar wind and meteorite influx fail to account for the relatively high nitrogen abundances. They may be explained by the possibility that indigenous nitrogen is high in the lunar rocks, as indicated by the fine-grained basalt 49,23, or that solar wind or meteoritic nitrogen is retained to a greater degree than is carbon.

The possibilities for contamination from nitrogen in returned lunar samples appear to be greater than for carbon. Possible sources are the exhaust gases from the lunar module and the nitrogen atmosphere to which the fresh lunar rocks were exposed at the Lunar Receiving Laboratory.

To see if nitrogen or carbon could be easily desorbed from lunar fines, a split of sample 86,A was heated for 24 hours at 300°C in a flowing helium atmo-

sphere. No carbon or nitrogen appeared to be removed by this treatment. If contamination by atmospheric nitrogen has taken place, it is strongly bonded to the fines material.

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4 January 1970

Instrumental Neutron Activation Analyses of Lunar Specimens

Abstract. *Ten Apollo 11 specimens were divided into 24 samples. Sodium contents of 8 diverse specimens cluster tightly about 0.3 percent. Plagioclase separated from sample 10044 contains about 1.09 percent Na; barium is not enriched in the plagioclase separate. Contents of the rare earths are strikingly high, and relative abundances resemble those of calcium-rich achondrites or abyssal basalts but are depleted in Eu by factors of 2 to 3 and in La by about 20 percent. The plagioclase separate is enriched in Eu and pyroxenes and opaque minerals are Eu-depleted. Fine fractions of 10044 are about 20 to 40 percent richer in most rare earths (50 percent for Eu) than coarse fractions, probably because of the presence of small grains in which rare earths are markedly concentrated. "Microgabbro" 10045 is impoverished, relative to the soil, in rare earths and Hf. Ratios by mass of Zr to Hf are comparatively low. Abundances of Mn, Co, Fe, Sc and Cr suggest systematic differences between igneous rocks on one hand and breccias and "soil" on the other. From the Co abundances, no more than about 3 percent of the present "soil" can consist of chondritic meteorite contamination.*

We have analyzed ten Apollo 11 specimens by techniques similar to those of Gordon *et al.* (1), with such modifications as recent experience dictates. Twenty-six elements were determined, but owing to space limitations we here report only salient observations and conclusions. A table of abundance and error estimates will be provided on request, and data and interpretations will be presented more fully in a subsequent paper (2).

Specimens were split where feasible to provide replicates. Two, 10044 and

10084, were treated more elaborately. The "microgabbro" powder, 10044, was split to yield a representative sample and then sieved with 100-mesh nylon screening. Fine material was split again, and the coarse material was hand-cobbed for separates of plagioclase, pyroxenes, and opaque minerals. The residue from the hand-cobbing, which remains representative of the coarse material to a good approximation, was split to give duplicate samples. The soil specimen, 10084, was sieved with 100-mesh and 325-mesh screening; splits of the origi-

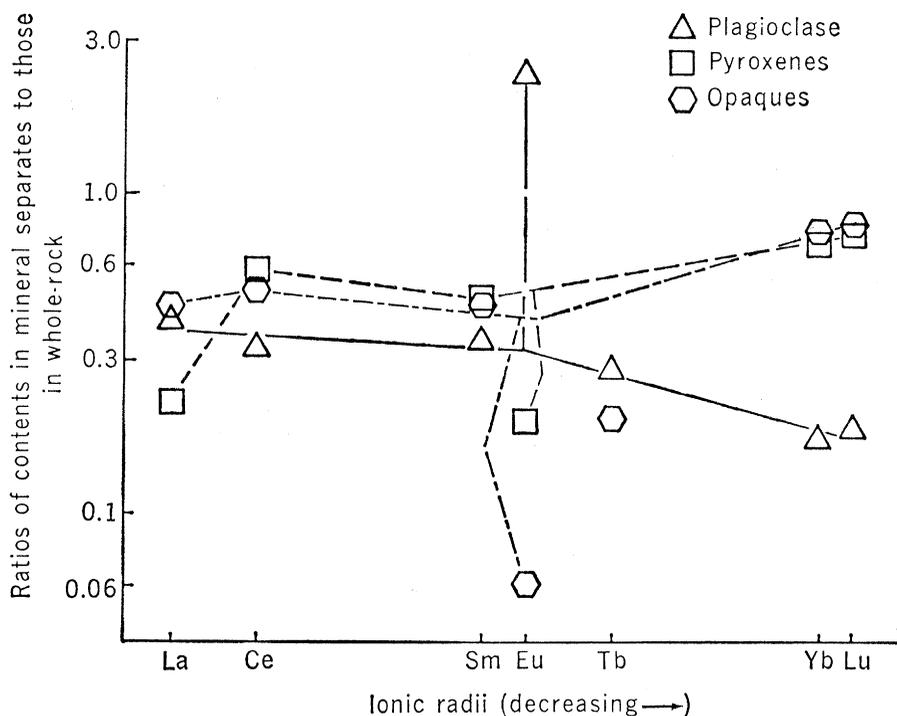


Fig. 1. Rare earth abundances in mineral separates from "microgabbro" 10044. Abundances have been corrected for cross-contamination as outlined in the text and then normalized to whole-rock rare earth contents. Logarithms (base 10) of normalized ratios are plotted versus ionic radii; ordinate values are those of the arguments of the logarithms. Whole-rock abundances would lie along the line at 1.0.

nal material and of the intermediate size-fraction were made.

Sodium abundances of bulk specimens are remarkably constant, averaging about 0.33 percent. All of the dispersion about the average value could be accounted for by analytical uncertainties and, in one case, sampling error. It seems unlikely that all specimens were derived from one or a series of closely related magmas, and in any case abundances of other elements do not display comparable constancy. One is led to suspect that some fundamental process, related perhaps to distribution coefficients for Na between solid and fluid phases, has limited the range of Na contents.

Before discussing distribution of elements among minerals separated from 10044, we must estimate contamination of one major "phase" by another (3). Sodium is a useful index of plagioclase contamination in the pyroxenes and opaque minerals separates; Al is even more useful in setting a limit on plagioclase in the opaque minerals. Plagioclase constitutes less than 6% of the pyroxenes, less than 2% of the opaque minerals. Chromium contents indicate that there is less than 1% of pyroxenes in the plagioclase, and Si contents limit pyroxenes to less than 15% in the opaque minerals separate. Less than 1%

opaque minerals are present in the plagioclase (Cr), less than 6% in the pyroxenes (Ti). The water-white plagioclase grains are the easiest to cobble cleanly, but it is not much more difficult to select pyroxene grains of high purity. It is difficult to detect silicates adhering to opaque grains under the microscope. Consequently, we shall assume that the plagioclase separate is contaminated with 0.5% of each of the other "phases", pyroxenes with 1% of each of the others, and opaque minerals with about 2% plagioclase and 5% pyroxenes.

Under these assumptions, Na contents of plagioclase, pyroxenes and opaque minerals are about 1.09%, 0.05%, and 0.016%, respectively. The latter two estimates are of course strongly dependent on the assumptions stated. Barium is not enriched in the plagioclase separate, within the precision of our data. Rare earth contents of the mineral separates are summarized, normalized to whole-rock abundances, in Fig. 1. Fractional crystallization of a combination of these major phases might leave residual magmas with relative rare earth patterns bowed upward in the middle of the sequence. We observe such patterns in our bulk specimens. Note the similarity of the plagioclase pattern, with a systematic decrease toward heavy rare earths and strong Eu

enrichment (Eu/Sm of 1.10 in contrast to 0.16 in the whole rock), to those of terrestrial and achondritic plagioclases (4). Both pyroxenes and opaque minerals are depleted in Eu (Eu/Sm of 0.067 and about 0.02). If the major rare-earth-bearing phases among the opaque minerals crystallized before plagioclases did (5), europic ion might well have been coherent with other trivalent rare earths in these phases. The Eu-depletion in the opaque minerals thus suggests that that element was present as europous ion at least early in the paragenesis. Depletion of Eu in pyroxenes might reflect crystallization in company with plagioclase, regardless of the oxidation state of Eu in the melt.

Contents of rare earths in bulk specimens are high. La contents range up to 26 ppm and Lu contents to 2.7 ppm. Relative abundances resemble those of Ca-rich achondrites or abyssal basalts (6). The contrast with terrestrial continental rocks, where high total rare earth contents are associated with relative enrichment of light rare earths, is striking. All lunar specimens are Eu-depleted by factors of two to three, from comparisons of Eu/Sm ratios with those of chondrites (7). This feature may be related to plagioclase fractionation or, more generally, to incoherence of europous ion and trivalent rare earths during magma formation. Lanthanum is also depleted (by about 10% to 30%) in comparison to values predicted from chondritic La/Ce ratios.

Rare earth abundances in sieved fractions of 10044 suggest that plagioclase is over-represented in the fine-grained fraction and that there are small grains in this rock in which rare earths are markedly concentrated. A crude estimate indicates that there is about 50% more plagioclase in the fine than in the coarse fractions. Independently, we note from Fig. 1 that as much as two-thirds of the total rare earths other than Eu may be present in minor (presumably late) phases or mineraloids, rather than in the major phases analyzed. Hafnium is distributed in a similar way: it is enriched in fine fractions of 10044, and a mass balance cannot be made using major phases alone. Note also that Hf is not concentrated in pyroxenes (unlike Sc), contrary to expectation.

The "microgabbro" 10045 is impoverished in rare earths and Hf relative to other igneous rocks, and even to breccias and the "soil". Rock 10045 resembles 10044 in these respects and the abundance anomalies may be related to

texture. The "soil", which presumably integrates across a wider range of rock types than represented in the Apollo 11 collection, seems to be weighted more toward rocks like 10044 and 10045 than toward 10022, 10071 and 10017. Breccias are not compositionally identical to the "soil" specimens analyzed, but are intermediate toward 10022, etc. Uranium contents range from 0.2 to 0.8 in whole rocks and in sieved fractions and mineral separates and tend to follow rare earths. These relationships should be viewed in the light of accurate geochronological data.

Ratios by mass of Zr/Hf range from about 25 to less than 13 and are low in comparison to terrestrial (and many meteoritic) values (8). We can at present offer no explanation for these remarkably low ratios. However, evidence for the dry and chemically reduced character of lunar magmas (5) suggests that terrestrial distribution coefficients and coherence relationships may not apply here.

Tantalum contents range from 1 to 2.3 ppm and are comparable to those in many continental crustal rocks (9). Tantalum is enriched in opaque minerals from 10044 relative to the whole-rock abundance.

Transition metal contents suggest that there are systematic differences between the igneous rocks, and the breccias and "soil". These trends perhaps display a negative correlation with those for the lithophilic elements noted above. From our Co abundances, one may show that no more than about 3% of the present "soil" can consist of chondritic fragments having Co contents like those of CL chondrites (10).

Both from data reported by the Preliminary Examination Team and from our results, it is clear that these rocks have been subjected to complex and unusual differentiation processes. Until we understand better the nature of these processes, it borders on metaphysical argument to compare the lunar rocks with, e.g., chondrites. Consequently, with the exception of a minor portion of our discussion of rare earths, we have eschewed such comparisons.

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4 January 1970

Isotopic Abundances of Actinide Elements in Lunar Material

Abstract. *The abundances of uranium and thorium were measured mass spectrometrically as 0.59 ± 0.02 and 2.24 ± 0.06 parts per million, respectively, in the fines of Apollo 11 bulk sample. The ratio $^{235}\text{U}/^{238}\text{U}$ was 0.007258 ± 0.000016 , in agreement with terrestrial uranium. The following upper limits were set: $^{236}\text{U}/^{238}\text{U} \leq 3 \times 10^{-9}$; parts per million of sample, $^{239}\text{Pu} \leq 1 \times 10^{-9}$; $^{244}\text{Pu} \leq 9 \times 10^{-11}$, and $^{247}\text{Cm} \leq 1.25 \times 10^{-10}$. More alpha activity from ^{227}Th and its decay products appears to be present than would be in equilibrium with the ^{235}U in the sample. The search for other actinide and transactinide nuclides is continuing.*

The purpose of our investigation was to measure the abundance and isotopic composition of thorium, uranium, and any transuranium elements that might also be present in lunar material, using mass spectrometric and radiometric techniques. Among the transuranium elements we made a specific search for ^{244}Pu and ^{247}Cm , two isotopes which had been produced synthetically and whose half-lives had been measured as $(8.18 \pm 0.26) \times 10^7$ (1) and $(1.64 \pm 0.24) \times 10^7$ years (2), respectively. The abundances of uranium and thorium not only contribute to the total distribution of the elements in lunar material, but are also an important ingredient in calculating the radioactive heat production on the moon. The possible presence of ^{244}Pu or ^{247}Cm , two extinct or nearly extinct nuclides, was interesting because they would have indicated an infusion from a supernova that occurred quite close to the solar system within the last billion years. Another object of our study was to examine the separated actinide and transactinide element fractions by alpha particle analysis and spontaneous fission counting to detect the presence of short-lived nuclides.

Earlier work on terrestrial samples (1) showed that it was necessary to work in a laboratory that had never

been used for heavy element chemistry; consequently, a new laboratory was equipped for the lunar analysis. All the glassware was carefully leached, and most of the reagents were distilled before use. In some cases special high purity reagents were used without further treatment, but they were tested for Th and U content before being used in the chemical separation procedure.

An alpha count of an 8-mg sample of the fines gave 0.036 ± 0.011 count/min. The single spontaneous fission event observed in 13 days of counting could have been due to counter background.

Two 1-gram samples of fines from the bulk sample (sample No. 10084,75) were dissolved, and known quantities of ^{230}Th (5×10^{-8} g), ^{233}U (7×10^{-8} g), ^{236}Pu (5×10^{-14} g), and ^{242}Cm (2×10^{-14} g) tracers were added. Before use, each of the tracers was carefully purified

Table 1. Micrograms (metal weight) of actinide element per gram of lunar fines.

Element	Sample		
	1	2	3
U	0.600 ± 0.018	0.582 ± 0.018	
Th	2.27 ± 0.06	2.21 ± 0.06	
^{244}Pu	$< 3 \times 10^{-10}$	$< 1 \times 10^{-10}$	$< 9 \times 10^{-11} *$
^{239}Pu	$< 1 \times 10^{-9}$		$< 1 \times 10^{-9} *$
^{240}Pu	$< 3 \times 10^{-10}$		
^{247}Cm	$< 1.25 \times 10^{-10}$		

* Preliminary results.