primordial radionuclide concentrations are similar to those of the lunar rocks, the total meteorite contribution to the lunar fines must not have been large enough to significantly alter its composition. The lunar materials thus appear rather unique in their primordial radionuclide composition, which indicates a substantially different geochemical history than that of known earth and other extraterrestrial materials.

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Elemental Compositions and Ages of Lunar Samples by Nondestructive Gamma-Ray Spectrometry

Abstract. *A gamma-ray spectrometry system with low background was used to determine the radioactivity of crystalline rocks, breccias, and fine material. Nuclides identified were* ^{40}*K,* ^{232}*Th,* ^{238}*U,* ^{7}*Be,* ^{22}*Na,* ^{26}*Al,* ^{44}*Ti,* ^{46}*Sc,* ^{48}*V,* ^{52}*Mn,* ^{54}*Mn, and* ^{56}*Co. Concentrations of K, Th, and U ranged between 480 and 2550, 1.01 and 3.30, and 0.26 and 0.83 parts per million, respectively. Concentrations of thorium and uranium were those of terrestrial basalts, while the potassium concentrations were near values for chondrites. Products of low-energy nuclear reactions showed pronounced concentration gradients at rock surfaces. Concentrations of K and of* ^{22}*Na determined here were combined with concentrations of rare gases to estimate gas-retention ages and cosmic-ray exposure ages with ranges of 2200 to 3200 and 34 to 340 million years, respectively, for three rocks.*

Lunar samples from Apollo 11 were analyzed by nondestructive, gamma-ray spectrometry at the Lunar Receiving Laboratory (LRL), Houston, Texas (*1*). The preliminary data have now been refined and additional samples also have been analyzed.

The low-background, gamma-ray spectrometer used in these studies has been described (*1*). Because the samples were viewed by two detectors, gamma-ray spectrums could be recorded either as noncoincident, or singles, data if an event occurred in one detector only, or as a gamma-gamma coincidence event if a signal was produced in both detectors simultaneously. The data acquisition system included a coincidence-anticoincidence logic circuit and dual 12-bit analog-to-digital converters interfaced to an on-line, digital computer. The computer was programmed to record singles data from each detector at a resolution of 256 channels, gamma-gamma coincidence data in a folded matrix with a resolution of 127 by 127 channels but requiring only 8192 storage locations, and a sum-coincidence spectrum of 256 channels. All of these spectrums were used in the detailed analysis of each sample.

It was necessary to use a variety of sample containers (three types of stainless steel and two types of polypropylene) with different attenuation and scattering properties for gamma rays. In addition, variations in radioactive contamination caused uncertainties in the background corrections. The preliminary examination was made especially difficult because it was not known in advance of the mission that some of these containers were to become necessary. Spectrum libraries for calibration in some cases had to be acquired during the active mission period, which reduced the time available for measuring lunar samples. After the sample quarantine was lifted, a number of small samples were measured which had been exposed to atmosphere, and so did not require special containers.

Before receipt of lunar samples, response functions of 19 expected nuclides were collected. Extended sources of electrolytically reduced iron powder containing 10^4 to 10^5 disintegrations per minute of each nuclide were prepared, measured, and compiled into a library of spectrums for the preliminary examination.

Potassium, thorium, and uranium standards were calibrated by weighing. Potassium standards were prepared from reagent-grade potassium chloride, and for some sources from potassium enriched in ^{40}K. Certified standards of thorium and uranium were obtained from the U.S. Atomic Energy Commission, New Brunswick Laboratory. All other radionuclides were standardized by absolute gamma-ray spectrometry to an overall uncertainty of less than 5 percent.

Final calibrations were performed through use of replicas filled with radionuclide standards which reproduced closely the bulk and electronic densities of the lunar samples. The replicas consisted of 0.08-mm aluminum-foil shells hand-formed around a plaster cast of each lunar sample, reinforced with epoxy cement and filled with iron powder. Electronic densities were calculated from the gross chemical composition (*1*) and matched in the replicated standards by use of a mass of iron powder about 5 percent in excess of the lunar sample mass. The bulk density was reproduced by the addition of 3-mm polyurethane spheres (0.016 g/cm^3). Such exact replicas were prepared for the four crystalline rocks measured. Lunar fines were simulated with mixtures of powdered iron and dunite. Nine replicas of each lunar sample were filled with a known portion of each radionuclide found in the preliminary examination. A spectrum library was collected for each set of replicas and used for final analysis of the five samples. The breccia samples were analyzed with cyclindrical standards used in the preliminary examination.

The concentration of each nuclide

Table 1. Gamma-ray analyses of whole rocks and fines from Apollo 11. Values for short-lived nuclides have been corrected for decay to 0000 hours, C.D.T., 21 July 1969. TI, tentatively identified.

Measurement	Sample number (type)*							
	10057 (A)	10072 (A)	10003 (B)	10017 (B)	10018 (C)	10019 (C)	10021 (C)	10002 (D)
Weight (g)	897	399	213	971	211.5	234	157	301.5
Bulk density (g/cm^3)	2.73 ± 0.14	2.37 ± 0.24	2.88 ± 0.35	3.00 ± 0.15	2.0 ± 0.2	2.02 ± 0.15		1.55 ± 0.05
K (ppm)†	2550 ± 130	2300 ± 120	480 ± 25	2430 ± 120	1420 ± 70	1200 ± 60	1600 ± 80	1100 ± 60
Th (ppm)†	3.30 ± 0.20	2.80 ± 0.17	1.01 ± 0.06	3.25 ± 0.18	2.30 ± 0.20	1.90 ± 0.19	2.50 ± 0.25	1.92 ± 0.10
U (ppm)†	0.79 ± 0.06	0.76 ± 0.06	0.26 ± 0.03	0.83 ± 0.07	0.60 ± 0.09	0.43 ± 0.06	0.54 ± 0.08	0.49 ± 0.04
^{26}Al (dpm/kg)	75 ± 8	73 ± 8	74 ± 8	73 ± 8	108 ± 16	101 ± 15	110 ± 15	120 ± 12
^{22}Na (dpm/kg)	41 ± 4	46 ± 5	41 ± 4	39 ± 4	55 ± 8	47 ± 7	55 ± 8	51 ± 5
^{44}Ti (dpm/kg)	TI	TI						TI
^{46}Sc (dpm/kg)	10 ± 2	8 ± 2	13 ± 3	13 ± 3	13 ± 4	10 ± 3	13 ± 4	8 ± 2
^{48}V (dpm/kg)			12 ± 9	11 ± 7				
^{52}Mn (dpm/kg)			35 ± 20				33 ± 21	
^{54}Mn (dpm/kg)	32 ± 6	20 ± 4	35 ± 7	33 ± 7	38 ± 10	28 ± 9	21 ± 6	28 ± 7
^{56}Co (dpm/kg)	31 ± 8	40 ± 10	43 ± 10	26 ± 7	33 ± 10	35 ± 10	50 ± 15	40 ± 7
^{7}Be (dpm/kg)	TI		TI	TI				TI

*Classification according to (*1*). † Standardization for assay of K, Th, and U with reference to terrestrial isotopic abundances. Equilibrium of Th and U decay series also assumed.

was determined from a resolution of the complex gamma-ray spectrum by the method of least squares (*2*) with the Univac 1108 computer complex at the Manned Spacecraft Center. Each of the four types of spectrums [singles from both NaI(Tl) detectors, sum coincidence, and two-parameter] were analyzed separately. Data from the two-parameter array were selected by adding adjacent planes in four regions of interest; the energy interval of each resulting spectrum was restricted to give the optimum discrimination against interferences.

In the preliminary analyses the extended sources described above were used to accumulate the spectrum libraries. When the counting sample did not match the particular reference library, an interpolation was required. The agreement between these preliminary results and the final results obtained with replicas was generally within 10 percent.

To check the experimental method, a test sample was prepared in a replica of sample 10017 containing K, Th, U, and ^{26}Al at the same concentrations found in the preliminary examination. The test sample was measured for a time similar to that employed during the acquisition of lunar sample data (1428 minutes). Analysis of the test sample on the basis of the exact library yielded observed concentrations that differed from the added concentrations by less than the statistical errors of the determination (0.9 percent for K, to 7 percent for ^{26}Al).

Because of the complex operations involved in handling lunar material, preparation of samples for gamma-ray spectrometry was slow during the quarantine period, and analysis of the first sample could not begin until 4 days after samples arrived at the LRL. Thus, radioactive species with half-lives of less than a few days were undetectable. In addition, the intense interferences from the gamma rays of the Th and U decay series in the samples made detection of weak gamma-ray components difficult. The precision of the measurements on these samples could have been improved by longer counting times; however, since these analyses were part of the preliminary examination at the LRL, time was limited. All samples were measured in at least two separate experiments, and total counting times were from 1000 to 3000 minutes per sample.

The results are summarized in Table 1. Twelve radioactive species were identified, some tentatively. The nuclides of shortest half-life identified were ^{52}Mn (5.7 days) and ^{48}V (16.1 days). The concentrations listed in Table 1 represent averages for the whole sample (usually a rock). Thus, if the sample is inhomogeneous, data obtained on a small portion may not be directly comparable to the concentrations of Table 1. The errors assigned in Table 1 include, in addition to the statistical errors of counting, estimates of possible systematic errors due to uncertainties in the detector efficiency calibrations.

Bulk densities for seven samples were obtained from the sample weights and the volumes of the replica shells fabricated as described above and are presented in Table 1. The crystalline rocks, breccias, and fines show a distinct correlation between bulk density and sample type.

The breccias and fines are very similar in their chemical compositions, as evidenced by their similar concentrations of K, Th, U, and cosmogenic radionuclides. This substantiates the hypothesis that the breccias are compacted mostly from the fine material. With the exception of sample 10003, the crystalline rocks appear to form

Table 2. Radionuclide concentrations in lunar samples as a function of depth. Concentrations for "Top" and "Interior" for 10017 were determined by an instrumental method and are only approximate. See text. Values for whole rock are from Table 1.

Nuclide	Sample 10017				Sample 10057	
	Top	Interior	Bottom	Whole rock	Top	Whole rock
^{26}Al (dpm/kg)	95 ± 15	65 ± 10	50 ± 7	73 ± 8	115 ± 9	75 ± 8
^{22}Na (dpm/kg)	43 ± 6	33 ± 6	30 ± 5	38 ± 4	63 ± 7	41 ± 4
^{56}Co (dpm/kg)	44 ± 18		10 ± 11	26 ± 7	90 ± 25	31 ± 8
K (% by weight)	0.235 ± 0.005	0.240 ± 0.005	0.240 ± 0.005	0.243 ± 0.012	0.258 ± 0.010	0.255 ± 0.013

Table 3. Estimates of gas-retention ages and cosmic-ray exposure ages for crystalline rocks. Gas contents from (*9*). Errors quoted for ages are combined analytical errors of gas contents and radioactivity determinations.

Sample	Gas content (10^{-8} cm^3/g)		Age (10^6 year)	
	^{40}Ar	^{21}Ne	K-Ar	Exposure
10017	4080 ± 200	47 ± 3	2200 ± 150	340 ± 37
10057	5200 ± 260	5.0 ± 0.3	2500 ± 180	34 ± 4
10072	7600 ± 380	25 ± 1	3200 ± 230	150 ± 17

a group with general chemical similarities. However, sample 10003, a coarsely crystalline rock resembling the terrestrial gabbro, differs significantly from the others in its concentrations of K, Th, and U. This suggests either that this rock came from another region of the moon or that it is the product of an igneous process different from that which formed the other crystalline rocks examined.

Potassium concentrations range from just below to three times the average potassium concentration of chondrites (0.085 percent). Such low K abundances are also characteristic of some oceanic gabbros and basalts (*3*). The Th and U contents are near those of terrestrial basalts, and the ratio of Th to U is constant at the terrestrial value of about 4. The abundance of radioactive elements (K, Th, and U) in the surface materials is much greater than that inferred for the mean content of radioactive elements in the moon (*4*), which suggests that the surface materials have been enriched in radioactive elements with respect to the whole planet. The ratio of K to U for the samples investigated here varies from about 1800 to 3200. This range of values is unusually low, both by comparison with chondrites (45,000) and with terrestrial rocks (10,000). Since this ratio is not readily changed by terrestrial igneous processes (*5*), it may be inferred that the average ratio (~2700) observed here does not vary greatly over the whole moon. This suggests that the moon, like the earth, is depleted in potassium relative to chondrites (*6*).

In addition to analyses of whole rocks, attempts were made to study radionuclide concentration gradients, since it was expected that solar flare protons could produce high concentrations of some nuclides at the surfaces of lunar materials (*7*). If nuclides decay by emission of coincident radiations, the presence of a concentration gradient on the surface of such an extended sample can be detected by variations in the intensities of the symmetrical energy zones of the two-parameter matrix. An instrumental method for deriving surface concentrations at upper and lower surfaces of a sample was developed, and a replica containing a steep surface gradient of ^{26}Al was used to derive a calibration factor. An exterior, 234-g portion of 10017 was counted in a special experiment to determine whether the surface had been exposed to solar proton bombardment. The area of the 1.46-Mev peak of ^{40}K was used as a guide for centering the sample between the detectors. These instrumental results, combined with conventional analyses of a fragment from the bottom of 10017 and a 7.6-g, 4-mm-thick specimen from the top of 10057, are shown in Table 2.

Although errors were large, concentration gradients were observed for ^{56}Co, ^{26}Al, and ^{22}Na. Because of the low threshold energy for the production of ^{56}Co, the concentration gradients for this nuclide are particularly steep. This suggests that solar proton activation is the principal source of the ^{56}Co, especially the flare (*8*) of 12 April 1969. The data of Table 2 suggest that ^{26}Al and ^{22}Na are produced both by solar and by cosmic-ray protons. Concentrations determined at the tops of the rocks agree approximately with values obtained for fine material. Since the fines were collected close to the surface, this agreement suggests that the instrumental method for distinguishing between the tops and bottoms of rocks is valid. The presence of such concentration gradients makes difficult a precise comparison between some data in Table 1 and data from fragments of the same rocks.

Information obtained by gamma-ray spectrometry may be combined with gas analysis data to provide estimates of crystallization and cosmic-ray exposure ages. In Table 3 are shown gas retention ages derived from the potassium analyses on three crystalline rocks (Table 1) combined with radiogenic ^{40}Ar concentrations obtained by Schaeffer *et al.* (*9*). Estimates of cosmic-ray exposure ages were made by the ^{22}Na-^{22}Ne method. From similar work on meteorites (*10*), it was assumed that the cross sections for the direct production of ^{22}Na and ^{22}Ne were equal. Specific activities of ^{22}Na from the present work were used in combination with Ne concentrations obtained by Schaeffer *et al.* (*9*). The ^{22}Ne of spallation origin was estimated (*9*) to be 10 percent higher than the concentration of spallogenic ^{21}Ne. The ages obtained are in good agreement with ^{3}He exposure ages determined by Schaeffer *et al.* (*9*) but are subject to considerable uncertainty in the production rates chosen for ^{22}Na and ^{22}Ne. Variations in cosmic-ray exposure ages of a factor of 10 are consistent with the concept of a continual series of disturbances of the lunar surface due to impact.

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