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Cosmogenic radionuclide concentrations and exposure ages of lunar samples from Apollo 12

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Abstract—Cosmogenic radionuclide abundances in a suite of samples from the Ocean of Storms were determined nondestructively by gamma-ray spectrometers of low background. Samples investigated were crystalline rocks 12002, 12004, 12039, 12052, 12053, 12054, 12062, and 12064; breccias 12013, 12034, and 12073; fines 12032 and 12070. The general concentration patterns of spallogenic radionuclides resemble those observed for Apollo 11 samples, but with some differences in detail. Cosmogenic radionuclides determined in this study were ^{22}Na, ^{26}Al, ^{46}Sc, ^{48}V, ^{52}Mn, ^{54}Mn, ^{56}Co, and ^{60}Co. Despite delays in obtaining samples during the preliminary examination, 5.7-day ^{52}Mn was determined in two rocks and 16-day ^{48}V was determined in four rocks.

Solar protons and galactic protons are both involved in the production of ^{22}Na, ^{26}Al, and ^{54}Mn in surface samples; however, several rocks show evidence of shielding. Concentrations of radionuclides in rock 12034 are consistent with production by galactic protons at depth, shielded from the effects of solar protons. Sample 12002,30 from the top of rock 12002 exhibited high concentrations of nuclides produced by solar flare protons, in confirmation of the orientation of 12002.

From the ^{60}Co concentration in rock 12002, a thermal neutron flux of 0.35 ± 0.18 neutrons cm^{-2} sec^{-1} was estimated. Estimates of cosmic-ray exposure ages were calculated by the ^{22}Na–^{22}Ne method. The results for seven samples are in good agreement with ^{3}He exposure ages by other investigators and range from 48 to 251 million years.

INTRODUCTION

THE EXTENSIVE studies of nuclides produced in the bombardment of meteorites by the solar and galactic cosmic rays have revealed much detailed information concerning the intensity and energy spectra of the incident radiations and their constancy with time. Lunar samples are even more suitable objects for such studies than meteorites, since the lunar samples have been irradiated in known orientations in space and are free from atmospheric ablation. A number of studies on radionuclide concentrations in Apollo 11 lunar samples (BEGEMANN *et al.*, 1970; HERZOG and HERMAN, 1970; O'KELLEY *et al.*, 1970a, 1970b; PERKINS *et al.*, 1970; SHEDLOVSKY *et al.*, 1970; WRIGLEY and QUAIDE, 1970) clearly demonstrated the potential of such information for elucidating the bombardment history of the lunar material, the histories of the incident particle fluxes, the erosion rates of rocks, and the rate of turnover of the lunar surface due to impact.

The absence of atmospheric ablation makes possible the detailed study of the effects of recent solar flares and long-term solar particle bombardment. Because of

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the short range of the solar particles and the high yields of some of the nuclear reaction products, gamma-ray spectrometry has been used quite successfully to determine the most recent orientation of rocks on the lunar surface (O'KELLEY *et al.*, 1970a, 1970b; PERKINS *et al.*, 1970; SCHONFELD and O'KELLEY, 1971).

Several radionuclides of interest have short half-lives. For this reason, much of the data reported below were recorded at the Lunar Receiving Laboratory (LRL), Houston, Texas, during the preliminary examination of the Apollo 12 samples. An early account of the results on some of the samples was given in LSPET (1970). Since the publication of the preliminary examination report the data analyses have been refined and further samples have been analyzed.

EXPERIMENTAL PROCEDURES

Several gamma-ray spectrometers were used in the course of this study. A NaI(Tl) scintillation coincidence spectrometer with an associated on-line, data acquisition system described by O'KELLEY *et al.* (1970b), together with a Ge(Li) spectrometer permitted rapid analyses of samples at the LRL during the quarantine period, so that nuclides of short half-life could be determined. Some studies at later times were carried out at Oak Ridge National Laboratory on a NaI(Tl) spectrometer similar to the scintillation spectrometer at the LRL.

The first Apollo 12 sample for radioactivity determination (12002,0) was received from the LRL Sample Laboratory on November 28, 1969, about 8.4 days after liftoff from the moon. During quarantine, samples were mounted in stainless steel containers for gamma-ray analysis. After quarantine, samples were generally sealed inside thin teflon bags for measurement. Methods of data acquisition and data analysis were essentially the same as those we used to analyze Apollo 11 samples.

For analyses of data on samples measured during the preliminary study, calibration of the LRL coincidence spectrometer was established by recording a library of spectra from cylindrical radioactive standards prepared by dispersing known amounts of radioactivity in quantities of iron powder. When recording the library of standard spectra, the standard sources were placed inside the steel containers actually used.

Spectrum libraries used for analyzing samples 12002,0; 12002,20; 12013,11; 12032,16; 12034,0; 12070,0; and 12073,0 were obtained from replicas which accurately reproduced the electronic and bulk densities of the lunar samples. Procedures for preparation of the cylindrical standards and the replicas were described earlier by O'KELLEY *et al.* (1970a, 1970b). A more detailed description of the analytical procedures employed for the Apollo 12 studies was given by O'KELLEY *et al.* (1971).

RESULTS AND DISCUSSION

Cosmogenic radionuclide concentrations

Our results on spallogenic radionuclides are given in Table 1. The general concentration patterns resemble those we observed in the Apollo 11 samples (O'KELLEY *et al.*, 1970a, 1970b); however, a number of subtle differences were noted due to effects of chemical composition and shielding. The data of Table 1 were recorded on large samples, usually a rock or a large fragment of a rock. Sample weights are listed in a companion paper by O'KELLEY *et al.* (1971). As observed in all gamma-ray spectrometry studies of Apollo 11 samples, the high concentrations of Th and U in lunar material makes difficult the determination of weak gamma-ray components. Because of the short times available for some of the measurements, it was not possible

Table 1. Concentrations (dpm/kg) of spallogenic radionuclides in Apollo 12 samples.* Values for short-lived nuclides have been corrected to 1426 GMT, Nov. 20, 1969.

Sample†	Type‡	^{22}Na	^{26}Al	^{46}Sc	^{48}V	^{52}Mn	^{54}Mn	^{56}Co	^{60}Co
12002,0	B	42 ± 3	75 ± 6	3.5 ± 1.0	13 ± 3	31 ± 12	38 ± 3	33 ± 4	0.55 ± 0.30
12002,20	B	47 ± 3	67 ± 5						0.73 ± 0.65
12002,30	B	86 ± 3	126 ± 6				50 ± 5	148 ± 20	
12004,1	A	53 ± 5	90 ± 6	3.7 ± 1.5			35 ± 4	34 ± 8	< 2.6
12039,0	B	43 ± 5	95 ± 7	< 6.0			37 ± 6	40 ± 10	
12052,1	A	40 ± 6	75 ± 6				27 ± 7	26 ± 10	
12053,0	A	40 ± 6	81 ± 12	7.0 ± 2.0	20 ± 5		35 ± 5	32 ± 6	< 1.0
12054,0	B	39 ± 7	50 ± 10	5.0 ± 2.0			36 ± 5	40 ± 10	< 1.0
12062,0	AB	30 ± 5	57 ± 9	5.0 ± 2.0	9 ± 3		31 ± 6	7 ± 4	
12064,0	B	40 ± 5	51 ± 5	5.0 ± 2.0	22 ± 6	33 ± 18	35 ± 3	32 ± 6	< 1.0
12013,0	C	50 ± 10	115 ± 16	< 15			< 66	50 ± 30	< 8.0
12013,11	C	26 ± 10	90 ± 10						
12034,0	C	29 ± 5	45 ± 5	< 10	< 60		16 ± 8	< 16	< 4.0
12073,0	C	63 ± 7	110 ± 10	< 10			28 ± 7	47 ± 12	
12032,16	D	48 ± 6	100 ± 7	< 10			27 ± 7	< 30	< 2.0
12070,0	D	70 ± 8	146 ± 16				41 ± 10	55 ± 14	< 1.5

* Upper limits are 2 σ evaluated from least-squares analysis.
† A zero following the 5-digit sample number designates a whole rock or fines sample.
‡ Petrologic type according to LSPET (1970).

to determine all 8 nuclides listed in Table 1 for all of the samples. Rock 12002 was the most carefully studied of all our samples and a rather complete radionuclide pattern was obtained. Except for some exceptions noted below, agreement within experimental error was obtained in the few cases where other radionuclide measurements on the same samples could be compared (RANCITELLI *et al.*, 1971; FINKEL *et al.*, 1971).

As was noted previously, ^{22}Na and ^{26}Al are produced both by solar and galactic cosmic rays (SHEDLOVSKY *et al.*, 1970; PERKINS *et al.*, 1970; O'KELLEY *et al.*, 1970a,b). Because the chemical composition of lunar material favors production of these nuclides, because they can be measured nondestructively by gamma-gamma coincidence methods with high sensitivity, and because their different half-lives (2.6 and 7.4×10^5 years) probe different regions of geologic time, their yields are of great interest.

The concentrations of ^{22}Na and ^{26}Al from Table 1 may be compared with calculated concentrations produced by galactic protons alone. This permits an estimate of the solar proton component. The ^{26}Al production in a 2π geometry was estimated by the method of FUSE and ANDERS (1969) and the ^{22}Na production was estimated by the method of BEGEMANN *et al.* (1970). Chemical compositions were taken from the best values from the Apollo 12 Lunar Science Conference and from the Apollo 12 Lunar Sample Catalog (WARNER, 1970).

The comparison between measured and calculated values is shown in Table 2. As a test of the calculations we show in Table 2 data on a sample 10017, ARA which was taken from the bottom of a well-oriented rock, as discussed by O'KELLEY *et al.* (1970b). This bottom piece from 10017 was shielded by about 14 g/cm^2 of rock, which effectively absorbed the solar protons. Agreement between calculation and experiment is good. Rock 12034 is a breccia recovered from a trench on the north rim of Head Crater; its burial depth was estimated as 15 cm (SHOEMAKER *et al.*, 1970). The low values for the concentrations of ^{22}Na and ^{26}Al obtained experimentally

Table 2. Comparison between measured concentrations of ^{26}Al and ^{22}Na in lunar rocks and fines compared with concentrations calculated for galactic production only

Sample	^{26}Al (dpm/kg)		^{22}Na (dpm/kg)		Remarks
	Measured	Calculated*	Measured	Calculated*	
Crystalline rocks					
10017,ARA	50 ± 7	41	30 ± 5	33	Bottom piece
12002,0	75 ± 6	42	42 ± 3	41	
12002,30	126 ± 6	42	86 ± 3	41	top slice
12004,1	90 ± 6	43	53 ± 5	40	
12052,1	75 ± 6	47	40 ± 6	37	
12053,0	81 ± 12	46	40 ± 6	37	
12062,0	57 ± 9	45	30 ± 5	36	
12064,0	51 ± 5	49	40 ± 5	35	
Breccias					
12013,0	115 ± 16	58	50 ± 10	41	
12034,0	45 ± 5	52	29 ± 5	40	buried 15 cm
12073,0	110 ± 10	50	63 ± 7	38	
Fines					
12032,16	100 ± 7	51	48 ± 6	41	
12070,0	146 ± 16	50	70 ± 8	40	

* Production in 2π geometry by method of FUSE and ANDERS (1969).
† Production rates estimated by method of BEGEMANN *et al.* (1970).

show that the rock was shielded from recent solar-proton bombardment. Consideration of the solar-proton spectrum (EBEOGLU and WANIO, 1966; LAL *et al.*, 1967) and the available information on variations in ^{22}Na and ^{26}Al concentrations with depth in lunar materials (FINKEL *et al.*, 1971; RANCITELLI *et al.*, 1971; ELDRIDGE *et al.*, 1971) conservatively specify a burial depth of $\gtrsim 8$ cm. Agreement between measured and calculated nuclide concentrations shown in Table 2 for 12034 is also good. It appears that the calculation of BEGEMANN *et al.* (1970) overestimates the ^{22}Na yields slightly.

Samples 12002,30 was a 46-g piece cut from the top of oriented rock 12002,0 and was investigated to obtain depth variations of cosmogenic nuclides by FINKEL *et al.* (1971). Before 12002,30 was submitted to destructive analysis, the data in Table 2 were obtained. As expected, high concentrations of ^{26}Al and ^{22}Na were seen, in excess of the production by galactic protons. It will be noted from Table 1 that the concentration of ^{54}Mn has also been enhanced over the nominal value by the solar proton bombardment while ^{56}Co which is almost totally produced by solar flares manifests a large surface concentration gradient. In contrast, breccia 12034 was shielded from solar protons and shows a low concentration of ^{54}Mn and undetectable ^{56}Co.

For the other rocks of Table 2 large excesses of ^{26}Al over that produced by galactic protons is observed, with moderate excesses of ^{22}Na. These results, together with the ^{56}Co concentrations of Table 1 show that the rocks in question were at least partially exposed on the lunar surface.

Rocks 12054, 12062, and 12064 show evidence of recent low exposure. Within experimental errors it is not possible to decide whether the low values of ^{26}Al are due to partial shielding from solar protons or whether the ^{26}Al did not attain saturation. It will be shown below that galactic proton exposure ages suggest that 12062 and 12064 have been near but not on the lunar surface for the last 150–200 m.y., which may indicate that these rocks received a low exposure to solar protons. Another possible explanation would be a high surface erosion rate, but it is difficult to understand why certain rocks erode rapidly while others do not.

It may be noted that for samples whose radionuclide concentrations can be compared (12062, 12034, 12070) our ^{22}Na concentrations agree with those of RANCITELLI *et al.* (1971) within experimental errors, but our ^{26}Al concentrations appear to be consistently lower.

The two soil samples we examined appear to have been taken from quite different depths. The high concentrations of ^{22}Na and ^{26}Al, and especially the high ^{56}Co, are consistent with near surface sampling for 12070. The sample of 12032 apparently came from a deeper zone, about 5 cm below the surface.

The concentrations of ^{46}Sc in samples from Apollo 12 are about 2.4 times lower than those we found in Apollo 11 samples. This reduction reflects the lower concentration of Ti target nuclei in the samples from the Ocean of Storms.

Despite delays in obtaining samples during the preliminary examination, 5.7-day ^{52}Mn was determined in two rocks and 16-day ^{48}V was determined in four rocks. The ^{52}Mn yields are approximately as expected from the chemical composition and correlate well with the ^{54}Mn yields. Most of the ^{48}V is produced by solar protons via the reaction ^{48}Ti(p, n)^{48}V. To correlate the observed ^{48}V yields with chemical composition, it was necessary to estimate the production of ^{48}V from spallation of iron by high-energy protons. This estimate was derived from the ^{48}V and ^{56}Co concentrations of rock 12062, which showed low exposure to solar-flare protons. By assuming that all ^{56}Co in 12062 was produced by solar-flare protons, the corresponding concentration of ^{48}V was estimated by use of the chemical composition in WARNER (1970) and the (p, n) cross sections for producing ^{48}V and ^{56}Co as measured by TANAKA and FURUKAWA (1959). Of the 9 dpm/kg of ^{48}V shown in Table 1 for 12062, about 3 dpm/kg could be attributed to solar flare production. The 6 dpm/kg of ^{48}V produced by high-energy spallation is not expected to vary significantly among the crystalline rocks of Table 1 because of the nearly constant concentration of iron.

In Fig. 1 we show that the yields of ^{48}V corrected to November 20, 1970, correlate well with the average titanium concentrations reported in the literature for 12002, 12053, and 12064. The flare responsible for the solar ^{48}V occurred on November 3, 1970; if the solar contribution is corrected to that date, the difference between the solid and dashed lines of Fig. 1 will be doubled.

Thermal neutron flux

Cobalt-60 has a half-life of only 5.3 years and is produced with a high cross section (37 barns) by thermal-neutron capture in ^{59}Co. Production of ^{60}Co either by spallation or by the (n, p) reaction in Ni is very low because of the small abundance of the target isotopes and the low cross sections for the nuclear reactions concerned. The concentration of ^{60}Co in lunar material can be employed to calculate the neutron flux characteristic of recent, steady-state production on the lunar surface. Such

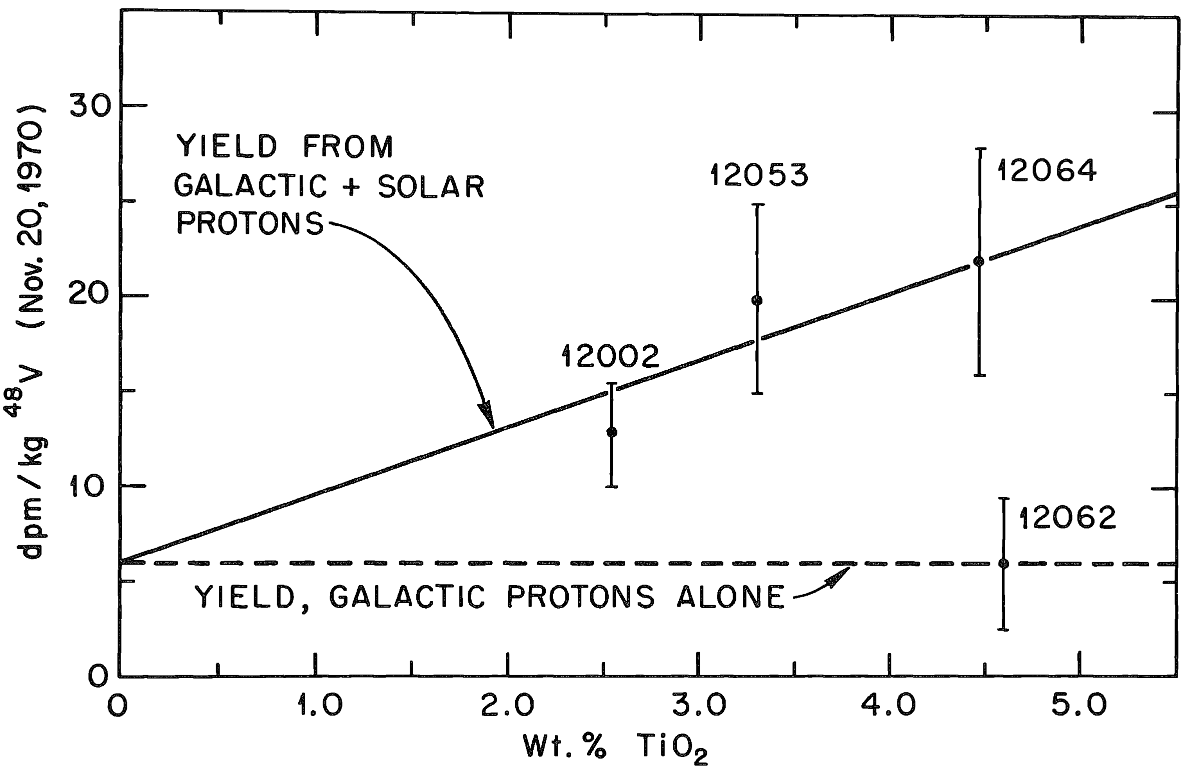

Fig. 1. Correlation between ^{48}V induced by solar-flare protons and titanium concentration. The solid line includes both the yield from solar protons calculated, as described in the text, and the yield from galactic protons.

information is a useful complement to fluxes deduced from mass spectrometric measurements of isotopic anomalies in Gd. The Gd isotope ratios yield an integrated thermal-neutron flux which requires a meaningful exposure age before an average flux can be obtained. Lunar rocks endure such a complex history that the average flux obtained mass spectrometrically may not represent the most recent flux to the accuracy desired.

Although ^{60}Co can be determined in lunar samples by gamma-ray spectrometry, rather large samples are required because the stable ^{59}Co target nuclide is present in such low concentration. Further, the intense interferences from abundant U and Th and cosmogenic radionuclides make difficult the resolution of small quantities of ^{60}Co.

In Table 1 we show that in the case of rock 12002,0 a value of 0.55 ± 0.30 dpm/kg was obtained for the ^{60}Co concentration. Based on an average Co concentration of 70 ppm in 12002, the thermal-neutron flux was found to be 0.35 ± 0.18 neutrons cm^{-2} sec^{-1}. The average mass density of rock 12002 was approximately 20 g/cm^2. Our result for a flux in a 20 g/cm^2 sample is in good agreement with the depth dependence of thermal neutron fluxes measured mass spectrometrically by Marti and Lugmair (1971) in lunar material of about 18 to 150 g/cm^2. Our result for 12002 is also in agreement with the theoretical value of 0.23 ± 0.06 neutrons cm^{-2} sec^{-1} calculated by Armstrong and Alsmiller (1971), who averaged the solar maximum and minimum fluxes and included nominal Apollo 11 rare-earth concentrations in the lunar surface composition.

Exposure ages

Estimates of cosmic-ray exposure ages were made by the ^{22}Na–^{22}Ne method as discussed by O'Kelley *et al.* (1970a, 1970b). It was assumed that the effective cross sections for production of ^{22}Na and ^{22}Ne were equal. Concentrations of Ne were obtained from the literature. The spallogenic ^{22}Ne was estimated to be 1.10 ^{21}Ne. Radioactive concentrations of ^{22}Na were taken from Table 1 and corrected for excess ^{22}Na of solar origin by a semiempirical factor.

In Table 3 we compare our exposure ages from the ^{22}Na–^{22}Ne method with ^{3}He exposure ages. The ^{3}He exposure ages were calculated from a production rate of 10^{-8} cm^3 STP ^{3}He/g per 10^6 years exposure. The agreement in Table 3 is gratifying and suggests that the ratio of production rates assumed for the ^{22}Na–^{22}Ne method is substantially correct.

The rocks of relatively shorter exposure age (12002, 12004, 12013, 12053) all were collected (Sutton and Schaber, 1971) in the Ocean of Storms north of a line connecting the north rim of Bench Crater and the center of Surveyor Crater. Rocks 12062, 12064, and 12065 have significantly longer exposure ages and were collected south of this line, which appears to be a boundary associated with Middle Crescent Crater. As shown by Warner and Anderson (1971), most of the crystalline rocks north of this diffuse boundary are porphyritic basalts, while those to the south are granular and ophitic basalts. The model proposed by Warner and Anderson to account for this distribution tentatively associated with Middle Crescent Crater suggests that the area north of the boundary would be strewn with ejecta of somewhat more recent exposure than the region to the south, which might be rich in older regolith material. Although this conclusion is speculative and is based on relatively few exposure ages, our data lend qualitative support to the Warner and Anderson model.

Table 3. Estimation of exposure ages of Apollo 12 lunar samples.

Sample	Exposure age (10^6 y)		Ref. gas data
	^{3}He	^{22}Na–^{22}Ne	
12002	89	96	a
12004	61	58	b
12013	40	48	c
12053	79	99	b
12062	150	153	b
12064	205	251	b
12065	182	217	b

a. ^{3}He from Hintenberger *et al.* (1971); ^{21}Ne from Marti and Lugmair (1971).
b. ^{3}He and ^{21}Ne concentrations from Hintenberger *et al.* (1971).
c. ^{3}He and ^{21}Ne concentrations from Schaeffer *et al.* (1970).

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