N72-22812

RADIOACTIVITIES IN RETURNED LUNAR MATERIALS

Semiannual Progress Report No. 2

For the period 1 August 1971 through 31 January 1972

Grant NGR 09-015-145

February 1972

Prepared for

National Aeronautics and Space Administration
Manned Spacecraft Center
Houston, Texas 77058

Smithsonian Institution
Astrophysical Observatory
Cambridge, Massachusetts 02138

RADIOACTIVITIES IN RETURNED LUNAR MATERIALS

Semiannual Progress Report No. 2

For the period 1 August 1971 through 31 January 1972

Grant NGR 09-015-145

February 1972

Prepared for

National Aeronautics and Space Administration
Manned Spacecraft Center
Houston, Texas 77058

Smithsonian Institution
Astrophysical Observatory
Cambridge, Massachusetts 02138

202-032

ABSTRACT

The H^3, Ar^{37}, and Ar^{39} radioactivities were measured at several depths in the large documented lunar rocks 14321 and 15555. The comparison of the Ar^{37} activities from similar locations in rocks 12002, 14321, and 15555 gives direct measures of the amounts of Ar^{37} produced by the 2 November 1969 and 24 January 1971 solar flares. From the Ar^{37} measurements, the intensities of the 2 November 1969 and 24 January 1971 solar flares were estimated to be $(5.1 \pm 1.2) \times 10^6$ and $(5.9 \pm 1.0) \times 10^6$ protons (> 50 Mev)/cm^2 sr, respectively. Because of the large differences in the K and Fe contents of the documented rocks, the Ar^{39} produced from the separate target elements, Fe + Ti, K, and Ca, at 1-, 5-, and 12-cm depths was obtained from the Ar^{39} measurements. The high-energy proton flux (> 200 Mev) averaged over 1000 yr was obtained from the Fe + Ti $\rightarrow$ Ar^{39}; the neutron flux ($\gtrsim$ 1 Mev) averaged over 1000 yr was obtained from the K $\rightarrow$ Ar^{39}; and the neutron flux ($\gtrsim$ 10 Mev) averaged over 1000 yr was obtained from the Ca $\rightarrow$ Ar^{39}. From the depth dependence of Ar^{39}, the intensity of solar-flare protons (> 50 Mev) averaged over the past 1000 yr was estimated to be $7 \times 10^8/cm^2$ yr. The tritium contents in the documented rocks decreased with increasing depth. The solar-flare intensity averaged over 30 yr obtained from the tritium depth dependence was approximately the same as the flare intensity averaged over 1000 yr obtained from the Ar^{39} measurements. Radioactivities in two Apollo 15 soil samples, H^3 in several Surveyor 3 samples, and tritium and radon weepage were also measured.

Radioactivities in Returned Lunar Materials

Grant NGR 09-015-145

1. INTRODUCTION

The radioactivities of Ar^{37}, Ar^{39}, and H^3 were measured in a number of Apollo 11 and 12 samples (Fireman et al., 1970; D'Amico et al., 1970, 1971; Stoenner et al., 1970a, b, 1971; Begemann et al., 1970; Bochsler et al., 1971). In the main, the measurements from different laboratories agree. D'Amico et al. (1971) measured the depth variation of Ar^{37}, Ar^{39}, and H^3 in rock 12002, a 6.4-cm-thick crystalline rock.

With samples from known locations in the large documented rocks, 14321 and 15555, we extended the Ar^{37}, Ar^{39}, and H^3 in a number of important ways. Apollo 14 material was subjected to a large solar flare on 24 January 1971, 2 weeks before sample recovery. No large solar flares occurred for several months before the Apollo 15 mission. The difference between the Ar^{37} activities from similar locations in 12002 and 15555 gives direct measures of the Ar^{37} activities produced by the 2 November 1969 flare, and the difference between the Ar^{37} activities in 14321 and 15555 gives the Ar^{37} activities produced by the 24 January 1971 flare. By use of measured Ar^{37} cross sections in simulated lunar material, the intensities of these flares were determined.

The target elements important for Ar^{39} production are Fe, Ti, K, and Ca. Rock 14321 is a breccia with low Fe and high K contents; rocks 12002 and 15555 have high Fe and low K contents; the Ca and Ti contents of the rocks differ only slightly. Since Ar^{39} is produced by the action of high-energy protons (> 200 Mev) on Fe and Ti, by low-energy neutrons ($\lesssim$ 1 Mev) on K, and by intermediate-energy neutrons ($\lesssim$ 10 Mev) on Ca, the Ar^{39} measurements from similar depths in the rocks determine the Ar^{39} from the separate target elements and the fluxes of the corresponding particles that interact with the separate target elements. The Ar^{39} has a 270-yr half-life so that the fluxes integrated over approximately 1000 yr are obtained from these determinations. The average high-energy proton flux for 1000 yr can be compared with measurements of the galactic rays; the neutron fluxes averaged over 1000 yr can be compared with the neutron fluxes that are obtained from Ar^{37} measurements.

The tritium was released in a two-step heating process, 3-hr heating at 275°C and melting, in the Apollo 14 and 15 and Surveyor 3 materials. The tritium released at 275°C may be either tritium implanted by the solar wind as suggested by D'Amico et al. (1971) or terrestrial contamination as suggested by Bochsler et al. (1971). The tritium released at high temperatures is mainly caused by solar-flare and galactic cosmic-ray interactions. The depth dependence of the tritium in rocks 14321 and 15555, Apollo 15 soil samples, and Surveyor 3 samples and its temperature-release pattern give information about the sources of the tritium and information about the intensity of solar flares integrated over the past 30 yr.

2. SAMPLE DESCRIPTION

Both brecciated rock 14321 and crystalline rock 15555 are documented rocks, which means that their position and orientation on the lunar surface were determined from photographs taken by an astronaut on the lunar surface. We received samples 14321, 81,* 14321, 267,† and 14321, 95 from rock 14321. A drawing of 14321, together with a slice taken from the center of the rock showing the locations of the samples, is given in Figure 1. Sample 14321, 81 extended from the top surface to 1.5-cm depth; the material from this sample was broken into three 0.5-cm-thick sections for separate analysis. The orientations of the sections were approximately preserved. Sample 14321, 95 was from the bottom of the rock at approximately 12 cm or 40 g/cm^2 depth. Sample 14321, 267 was from 5-cm depth. Although samples 14321, 81 and 14321, 95 were received 78 days after the Apollo 14 mission, sample 14321, 267 was received more than 6 months after the mission. Nearby samples of 14321 labeled 1, 2, 3, and 5 were analyzed by Wahlen et al. (1972) for a number of radioactive nuclides. Figure 1 was obtained from Wahlen et al. (1972). Rock 14321 was very fragile, so that samples could be broken with small hand tweezers without much difficulty.

Our samples of 15555 were taken from slice 15555, 57 shown by solid lines in Figure 2, where the outline of rock 15555 is shown by dashed lines. This drawing was made by members of the Lunar Receiving Labotatory. Sample 15555, 98 was a top-surface sample approximately 0.8 cm thick. Sample 15555, 80 was from near the middle of an adjacent column cut from slice 15555, 57 at approximately 6-cm depth, and sample 15555, 77 was from near the bottom of the same column at approximately 14-cm depth. These samples were received 119 days after the Apollo 15 mission. Two soil samples, 15271, 17 and 15261, 14, of 1.97- and 2.04-g weight were received 51 days after the mission; their Ar^{37} activities could be measured more accurately than those in the larger 15555 rock samples. Soil 15271, 17 was taken from the compressed wheel track of the lunar rover, and soil 15261, 14 was collected from a trench dug into the rim of a 12-m-diameter crater. The depth of the trench was estimated at approximately 30 cm.

---

*Samples 14321, 81 and 14321, 83 were combined and called sample 14321, 81.

†Samples 14321, 261; 14321, 262; and 14321, 267 were combined and called 14321, 267.

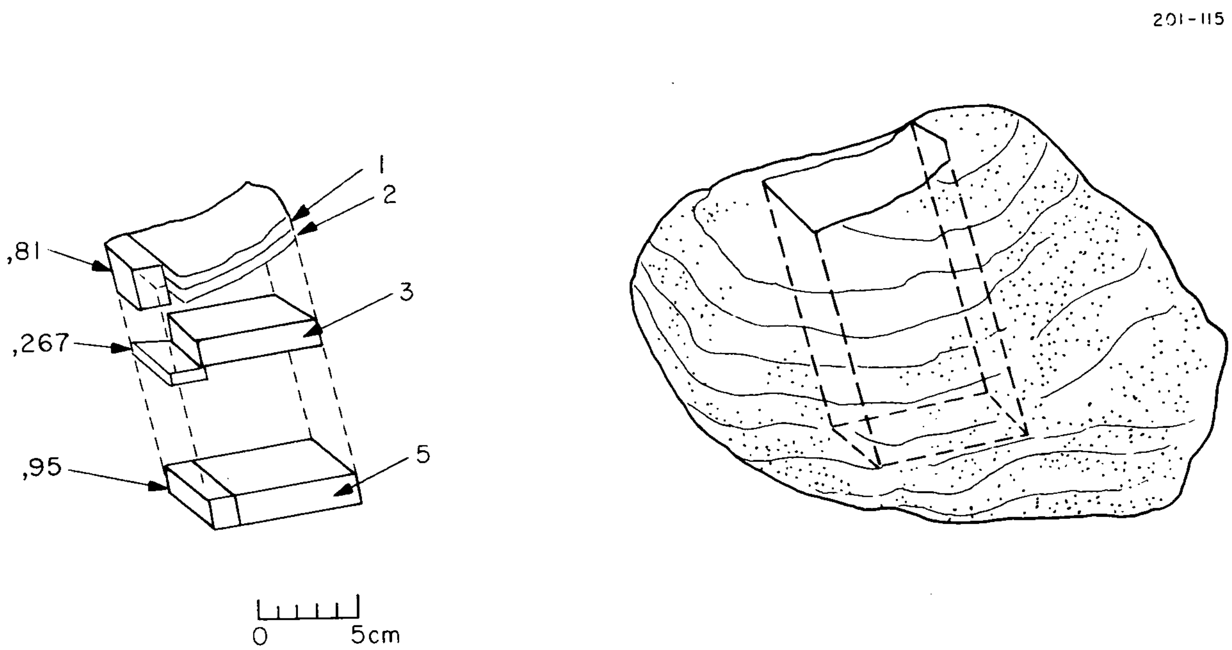

Figure 1. Lunar rock 14321 and sample locations.

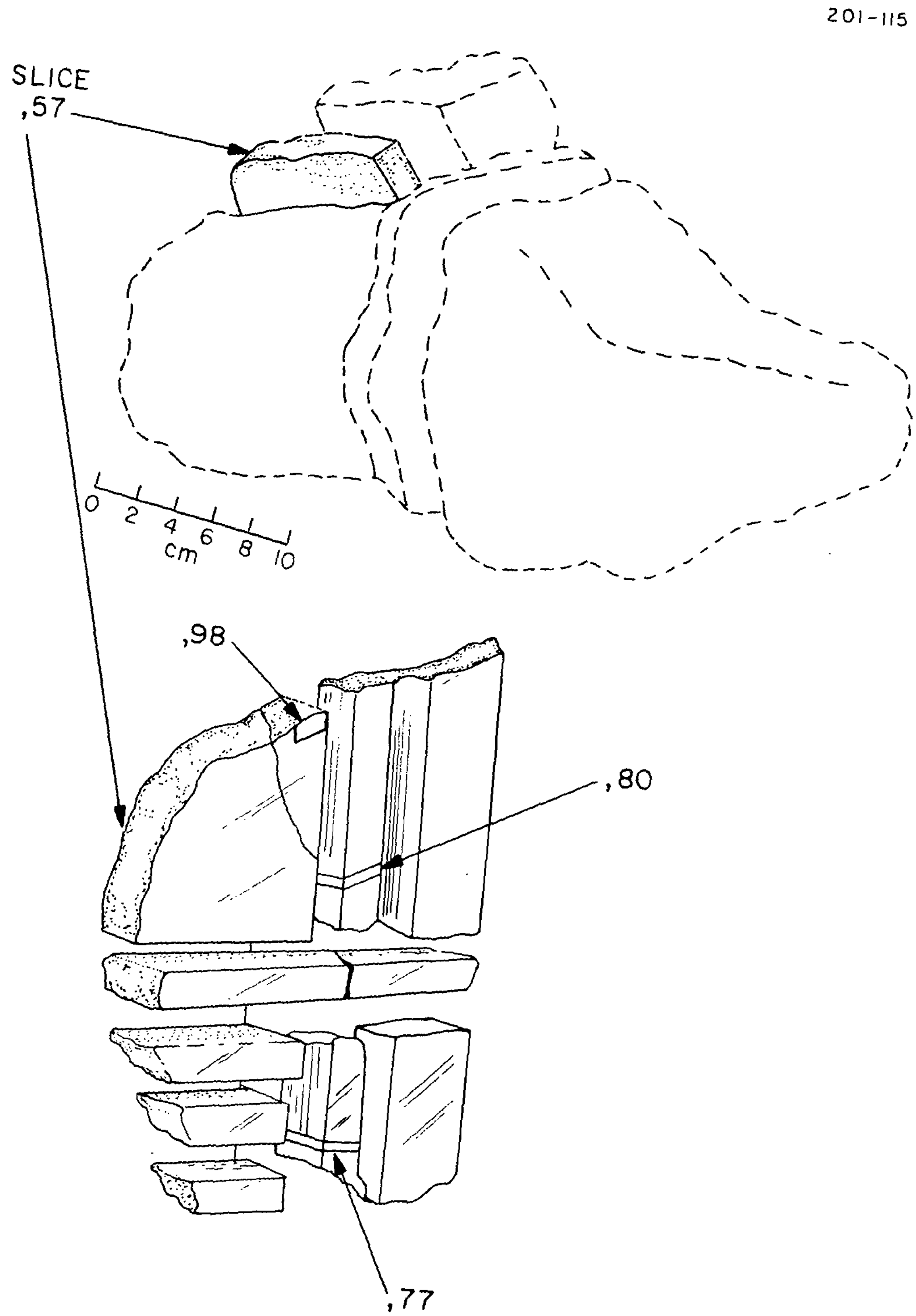

Figure 2. Lunar rock 15555 and sample locations.

3. EXPERIMENTAL PROCEDURE

Since Ar^{37} decays with a 35-day half-life, we measured the argon radioactivities as rapidly as possible and stored the tritium, except for that from the remelting of the samples, for later analysis. The tritium from the remelt was measured before proceeding to the next sample to ascertain that all tritium had been removed from the sample. The gamma-ray analysis of Apollo 14 samples done at LRL indicated that their uranium and thorium contents were 10-fold higher than those in most previous samples. We therefore put an additional charcoal trap in the purification system to ensure the complete removal of the larger amounts of radon expected from 14321 samples. Otherwise, the extraction and purification procedures for the gases from the 14321 samples were identical to those used for most previous lunar samples (D'Amico et al., 1971). The additional charcoal trap was not used in the processing of the Apollo 15 samples.

The gas was removed in three heatings. After the sample had been placed in a molybdenum crucible that had been previously outgassed, the extraction system was pumped down to a pressure of several torrs. Hydrogen carrier was added, and the sample was heated to 275°C with a resistance heater for 2 hr. The gases were removed with an automatic toepler pump and stored in a glass bulb for later purification and tritium analysis. The sample was then melted by induction heating in the presence of argon and hydrogen carrier. The gases from the molten Apollo 14 samples were collected on charcoal at liquid-nitrogen temperature and removed at dry-ice temperature, and then transferred to finely divided vanadium powder at 800°C. The gases from the molten Apollo 15 samples were transferred to the vanadium directly. The hot vanadium removed the chemically active constituents; the vanadium was then slowly cooled to room temperature to absorb hydrogen as vanadium hydride. The gas was then removed from the cool vanadium, and its volume measured. If the volume of gas was larger than the amount of argon carrier used, the gas was repurified over vanadium. This procedure was repeated until the volume of gas did not change. At this stage, the volume was the same (within several percent) as that of the argon carrier used. This gas was repurified over hot titanium and condensed on charcoal at liquid-nitrogen temperature. The argon was removed from the charcoal at dry-ice temperature and placed in the same small low-level proportional counters used for Apollo 11 and 12 samples (Fireman et al., 1970; D'Amico et al., 1970, 1971). Methane (10%) was added to the argon. The counter was removed from the purification system and counted in the low-level system, where the counts between 0.4 and 8.0 kev were recorded on a 100-channel analyzer, and those above 7.4 kev, on scalers. Figure 3 is a plot of the argon counting data for sample 14321, 95 for an 8-day period from 27 April to 6 May 1971. Figure 4 is a plot of the argon counting data for 15555, 80 for a 22-day period from 6 to 28 December 1971.

The hydrogen was recovered from the vanadium by reheating the vanadium to 800°C and pumping the released hydrogen into a storage bulb.

The sample was then remelted by induction heating, and wall deposits were heated with a torch in the presence of hydrogen carrier. The gas was transferred to finely divided vanadium powder at 800°C, which was slowly cooled to room temperature until no gas remained. The hydrogen was removed from the vanadium by reheating and passed through a charcoal trap at dry-ice temperature to remove any possible radon. After its volume had been measured, the hydrogen was added to a low-level proportional counter that contained 400-torr pressure of P-10 gas. The resolution of the counter was checked with an Fe^{55} source, and the counting was done in a low-level system.

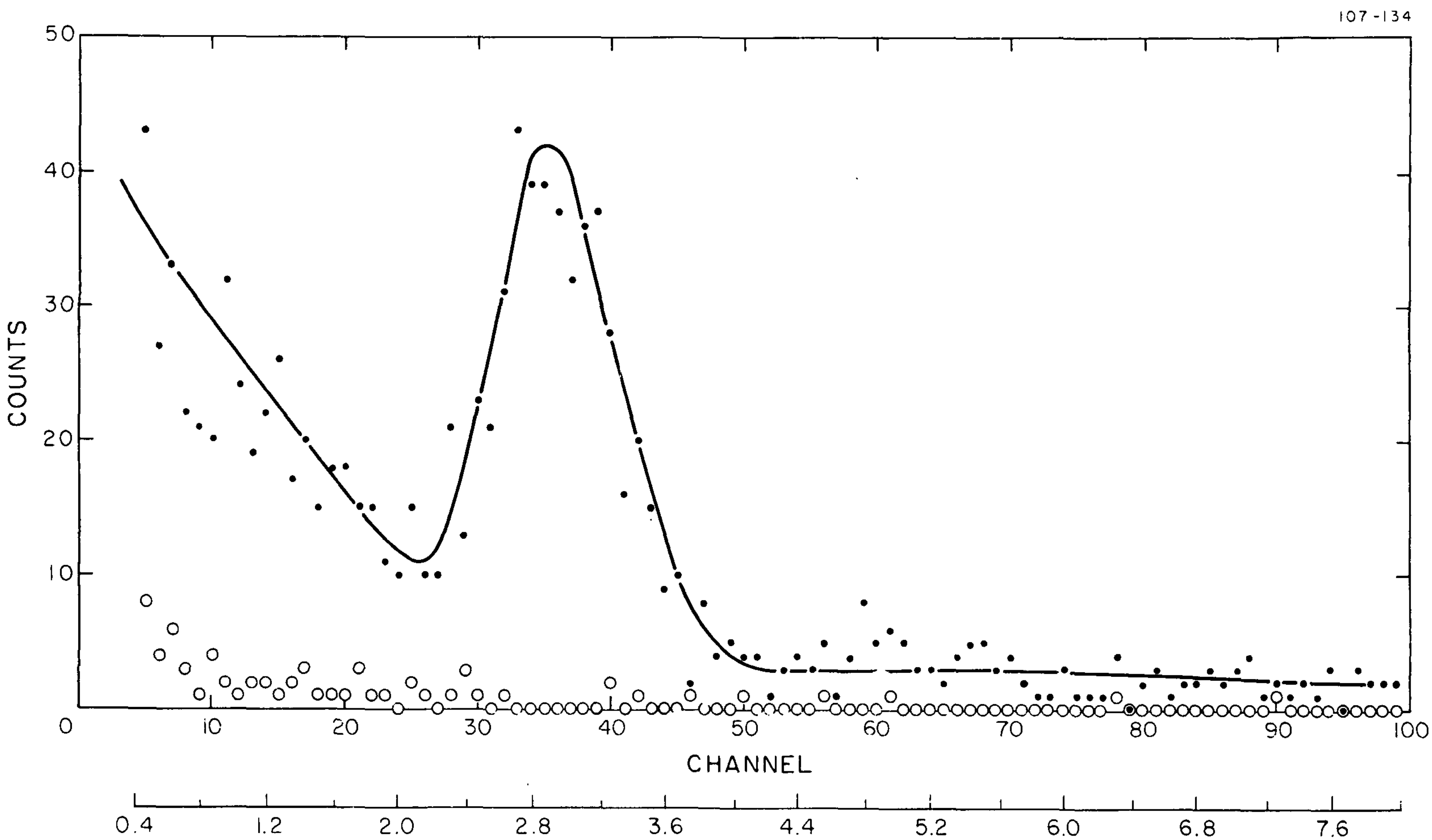

Figure 3. Argon radioactivities for 10.26 g of lunar rock sample 14321, 95. Curve is from calibrations with Ar^{39} = 14.8 ± 1.1 dpm/kg and Ar^{37} = 32.6 dpm/min. ● = counts/channel, 12158 min, 80 to 88 days after collection on 6 February 1971; counts beyond channel 99 = 157. ○ = background counts/channel, 11426 min; counts beyond channel 90 = 11.

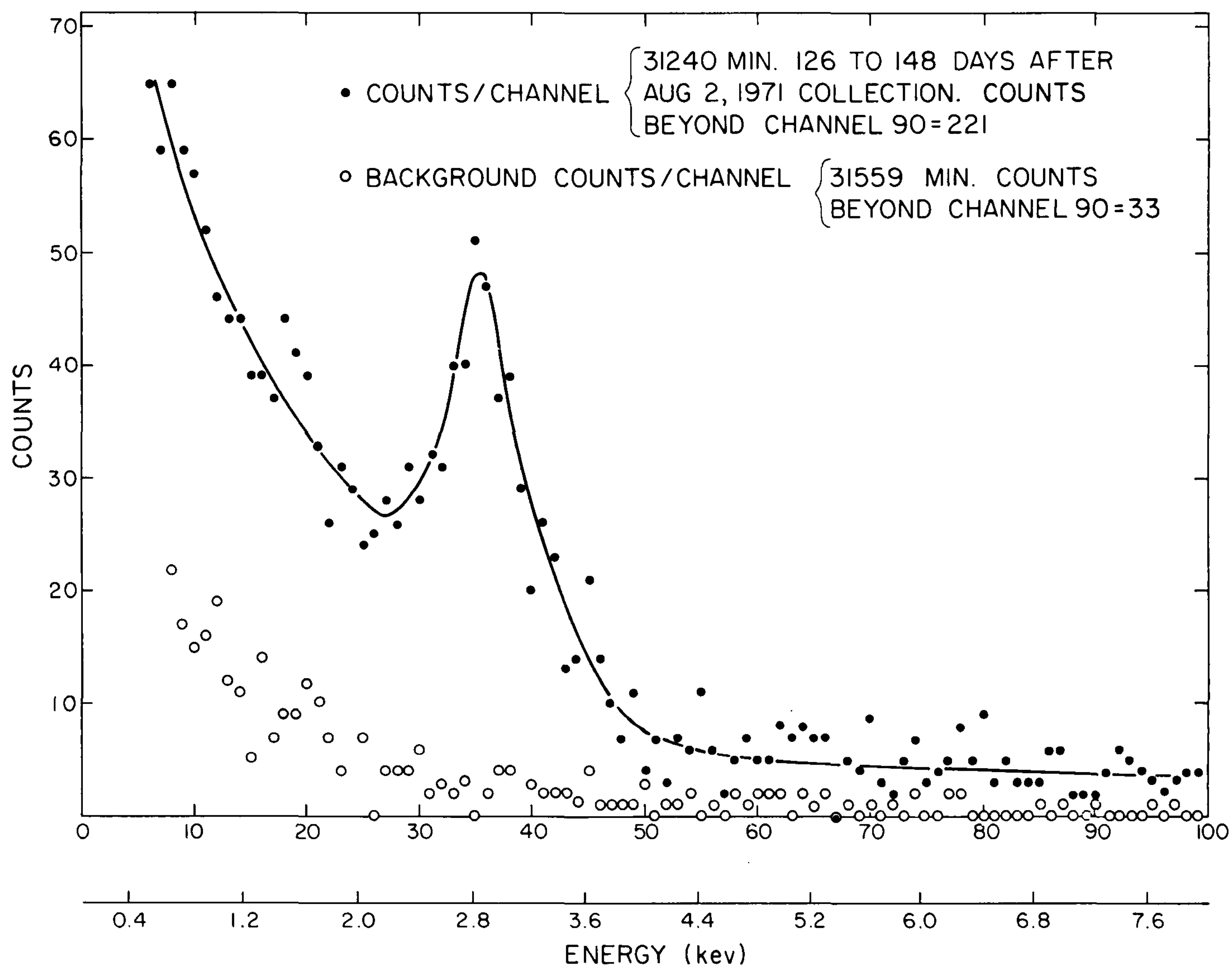

Figure 4. Argon radioactivity for lunar rock sample 15555, 80 (10. 25 g).

4. RESULTS

Table 1 gives the Ar^{37} activities that we have measured and the dates of large solar flares that occurred within 4 months before the missions. Apollo 15 samples were not exposed to any such flares; Apollo 14 samples were exposed to the 24 January 1971 flare 13 days before recovery; and Apollo 12 samples were exposed to the 2 November 1969 flare 21 days before recovery. The Ar^{37} activity in the top sample of 15555, 15.8 ± 3.0 dpm/kg is a factor of 2.5 lower than in the top sample of 14321 and a factor of 2 lower than in the top sample of 12002. The Ar^{37} activities in 15555 are produced solely by galactic cosmic rays; those in 14321 and 12002 are produced by both galactic cosmic rays and a solar flare. Ar^{37} activity is produced mainly by the action of protons and neutrons on Ca. The Ar^{37} activity per kg of Ca in the top sample of 15555 is slightly lower than in soils 15271, 10017, and 10084, showing that these soil samples, although not affected by solar flares, were taken from slightly larger average depths than sample 15555, 98 was. The solar-flare-produced Ar^{37} in the top samples of 14321 and 12002 were 364 ± 60 dpm/kg Ca and 324 ± 70 dpm/kg Ca, respectively; however, at approximately 6-cm depth, the solar-flare-produced Ar^{37} is only 55 ± 65 dpm/kg Ca.

Reedy and Arnold (1972) calculated the Ar^{37} activities as a function of depth by galactic cosmic rays on lunar material. The Ar^{37} activity measured at three depths in rock 15555 agrees quite well with Reedy and Arnold's calculations. It is not necessary to use Reedy and Arnold's calculations to obtain the intensities of the 24 January 1971 and the 2 November 1969 flares from the Ar^{37} measurements. D'Amico _et al_. (1971) measured the Ar^{37} production cross sections in a series of targets of simulated lunar material containing 6.7% Ca bombarded by 158-Mev protons. Interspersed between the lunar-type targets were iron absorbers so that the Ar^{37} production cross sections for 50-, 85-, and 158-Mev protons could be determined. The protons of these energies in the flares should contribute more Ar^{37} than those of higher energies if the differential energy spectrum varies as E^{-3}; the Ar^{37} production cross section, $\sigma_{Ca}(Ar^{37})$, is 60 ± 3 mb for 158- and 85-Mev protons and 10.8 ± 1.0 mb for 50-Mev

Table 1. Ar^{37} activities and solar flares within 4 months of mission.

Sample	Depth (cm)	Recovery date	Flare date	Ar^{37*} (dpm/kg)	Ca (%)	Ar^{37} (dpm/kg Ca)	$\Delta Ar^{37\dagger}$ (dpm/kg Ca)	Solar-proton flux ($1/cm^2$ sr)	Solar-proton energies
15555, 98	0–0.8	2 August 1971	none	15.8 ± 2.8	6.7	236 ± 42	–	–	–
15555, 80	~ 6	2 August 1971	none	31.2 ± 3.3	6.7	465 ± 49	–	–	–
15555, 77	~ 14	2 August 1971	none	35.0 ± 6.5	6.7	520 ± 97	–	–	–
14321, 81, 83	0–0.5	6 February 1971	24 January 1971	38.5 ± 2.5	6.4	600 ± 39	364 ± 60	$(5.9 \pm 1.0) \times 10^6$	(> 50 Mev)
14321, 81, 83	0.5–1.0	6 February 1971	24 January 1971	37.6 ± 2.5	6.4	590 ± 39	354 ± 60	–	–
14321, 81, 83	1.0–1.5	6 February 1971	24 January 1971	35.0 ± 4.0	6.4	550 ± 63	324 ± 75	–	–
14321, 95	~ 12	6 February 1971	24 January 1971	34.0 ± 2.7	6.4	530 ± 42	≤ 100	$(< 1.6) \times 10^6$	(>200 Mev)
12002, 57	0–0.8	23 November 1969	2 November 1969	30.0 ± 4.0	5.4	556 ± 65	320 ± 70	$(5.1 \pm 1.2) \times 10^6$	(> 50 Mev)
12002, 57	0.8–3.1	23 November 1969	2 November 1969	25.0 ± 1.5	5.4	463 ± 28	160 ± 50	$(2.6 \pm 0.9) \times 10^6$	(> 70 Mev)
12002, 59	4.9–6.4	23 November 1969	2 November 1969	27.5 ± 2.5	5.4	510 ± 46	55 ± 65	$(0.9 \pm 1.1) \times 10^6$	(>100 Mev)
Soil 15271, 17	0–5	2 August 1971	none	21.4 ± 2.0	8.1	264 ± 25	–	–	–
Soil 15261, 14	~ 30	2 August 1971	none	26.4 ± 3.0	–	–	–	–	–
Soil 10017, 14	0–5	21 July 1969	none	21.0 ± 2.0	7.4	284 ± 27	20 ± 37	–	–
Soil 10084, 24	0–5	21 July 1969	none	27.2 ± 2.2	8.8	309 ± 25	45 ± 35	–	–

*Activity at time of recovery.

†Solar-flare-produced Ar^{37} from calcium, $\Delta Ar^{37} = (Ar^{37}/\%\ Ca) - [Ar^{37}(15)/\%\ Ca]$, where $Ar^{37}(15)$ is the Ar^{37} in an Apollo 15 sample at a comparable depth.

protons. The cross section falls steeply between 85 and 50 Mev; below 50 Mev, the cross section probably falls even more steeply. On the basis of these cross sections and the solar-flare-produced Ar^{37} from calcium, ΔAr^{37}, which is defined in Table 1, we estimate the solar-flare-proton fluxes given in Table 1. The 24 January 1971 flare intensity is estimated to be $(5.9 \pm 1.0) \times 10^6$ protons/cm^2 sr greater than 50-Mev energy; the 2 November 1969 flare intensity is estimated to be $(5.1 \pm 1.2) \times 10^6$ protons/cm^2 sr (> 50 Mev). These flare intensities obtained from lunar rocks can be compared with satellite measurements. From the Explorer 35 satellite, Van Allen (private communication, 1972) obtained 4.9×10^6 protons (> 55 Mev/cm^2 sr) for the 24 January 1971 flare and 3.2×10^6 protons (> 55 Mev/cm^2 sr) for the 2 November 1969 flare with 20% uncertainty. The intensities of the flares obtained from the Ar^{37} activities in lunar rocks are slightly higher than those obtained by Van Allen from his Explorer 35 counters.

Table 2 gives the Ar^{39} activities at several depths in three oriented lunar rocks and in four soil samples. An interesting feature of the Ar^{39} activities is that the Ar^{39} increases with depth in 14321, a high K rock, but is approximately constant with depth in low K rocks 15555 and 12002. Ar^{39} is produced by high-energy protons on Fe (> 300 Mev) and on Ti ($\gtrsim$ 200 Mev); on the other hand, Ar^{39} is produced from K and Ca by the action of neutrons through the K^{39} (n, p) and Ca^{40} (n, 2p) reactions. The K^{39} reaction is exothermic; the Ca^{40} reaction has an 8.1-Mev threshold energy. The Ar^{39} production rates for four individual target elements can be calculated from Ar^{39} measurements in four lunar samples at the same depth; however, such calculated rates would have considerable error and there would be no consistency checks. If the Ar^{39} production rates from Fe and Ti are lumped together and the Fe + Ti $\rightarrow Ar^{39}$ production rate is estimated theoretically, then the K $\rightarrow Ar^{39}$ and Ca $\rightarrow Ar^{39}$ production rates are obtained from the data with relatively small errors and there are six samples that remain for consistency checks. We have therefore adopted this procedure for obtaining the Ar^{39} production rates in K and Ca.

In Table 2, two situations are represented: (1) the Ar^{39} from Fe + Ti is produced solely by galactic cosmic rays of the present average intensity and is given by Reedy and Arnold's calculation (private communication, 1972); and (2) extremely energetic

Table 2. Ar^{39} activities, estimated productions from target elements with and without energetic solar flares, and neutron fluxes.

							Situation 1: No solar-flare protons (> 200 Mev) past 1000 yrs			Situation 2: Solar-flare protons (> 200 Mev) past 1000 yrs equal galactic cosmic rays			Neutrons/cm^2 sec	
Sample	Depth (cm)	K (%)*	Fe (%)*	Ti (%)†	Ca (%)*	Ar^{39} (dpm/kg)	$Fe+Ti \rightarrow Ar^{39}$ (dpm/kg)†	$K \rightarrow Ar^{39}$ (dpm/kg)	$Ca \rightarrow Ar^{39}$ (dpm/kg)	$Fe+Ti \rightarrow Ar^{39}$ (dpm/kg)‡	$K \rightarrow Ar^{39}$ (dpm/kg)	$Ca \rightarrow Ar^{39}$ (dpm/kg)	> 1 Mev**	> 10 Mev††
15555, 98	0−0.8	0.025	17.5	1.36	6.7	8.4 ± 0.5	1.8	0.13	6.5	3.6	0.17	4.7		
15555, 80	~6	0.025	17.5	1.36	6.7	7.5 ± 0.5	1.7	0.3	5.5	3.4	0.5	3.7		
15555, 77	~14	0.025	17.5	1.36	6.7	9.8 ± 0.8	1.4	0.4	8.0	2.8	0.6	6.3		
14321, 81	0−0.5	0.37	8.6	1.2	6.1	8.8 ± 0.5	1.0	2.0 ± 0.6	5.8 ± 0.7	2.0	2.6 ± 0.6	4.2 ± 0.6	3.7	2.0
14321, 81	0.5−1.0	0.37	8.6	1.2	6.1	8.3 ± 0.7	1.0	2.0 ± 0.6	5.8 ± 0.7	2.0	2.6 ± 0.6	4.2 ± 0.6	3.7	2.0
14321, 81	1.0−1.5	0.37	8.6	1.2	6.1	9.5 ± 0.8	1.0	2.0 ± 0.6	5.8 ± 0.7	2.0	2.6 ± 0.6	4.2 ± 0.6	3.7	2.0
14321, 267	~5	0.37	8.6	1.2	6.1	12.1 ± 1.0	0.95	6.2 ± 1.1	5.0 ± 0.7	1.9	7.0 ± 1.2	3.2 ± 0.5	10.6	1.6
14321, 95	~12	0.37	8.6	1.2	6.1	14.8 ± 1.0	0.84	6.8 ± 1.2	7.2 ± 0.9	1.7	7.5 ± 1.3	5.7 ± 0.8	12	2.7
12002, 57	0−0.8	0.045	16.5	1.5	5.4	8.0 ± 0.7	1.8	0.3	5.2	3.6	0.4	3.7		
12002, 57	0.8−3.1	0.045	16.5	1.5	5.4	8.2 ± 0.5	1.8	0.4	5.2	3.5	0.5	3.7		
12002, 59	4.9−6.4	0.045	16.5	1.5	5.4	8.0 ± 0.6	1.7	0.7	4.4	3.4	0.9	3.0		
Soil 15271, 17	0−5	0.17	9.5	0.88	8.1	10.0 ± 1.5	1.0	0.9	7.7	2.0	1.2	5.6		
Soil 15261, 14	~30	−	−	−	−	7.1 ± 1.0	−	−	−	−	−	−		
Soil 10017, 14	0−5	0.24	15.3	7.1	7.4	16.4 ± 0.9	3.0	1.3	7.2	6.0	1.8	5.2		
Soil 10084, 24	0−5	0.11	12.3	4.4	8.8	12.1 ± 0.7	2.0	0.6	8.4	4.0	0.8	6.2		

*A weight percent obtained from various publications of chemical composition of lunar material.

†Reedy and Arnold's calculated galactic cosmic-ray production rate of Ar^{39} from Fe + Ti (private communication 1972).

‡Twice calculated galactic cosmic-ray production rate of Ar^{39} from Fe + Ti.

**Based on Ar^{39} from K^{39} with $\sigma(n, p) = 200$ mb.

††Based on Ar^{39} from Ca^{40} with $\sigma(n, 2p) = 40$ mb.

solar flares have occurred during the past 1000 yr, which raised the cosmic-ray intensity to twice the present average, so that the Ar^{39} from Fe + Ti is twice the Reedy and Arnold value.

The calculation proceeds by subtracting the Ar^{39} produced from Fe + Ti from the measured Ar^{39} to give the Ar^{39} from Ca + K. The Ar^{39} produced from Ca and the Ar^{39} from K are regarded as unknowns, and a pair of simultaneous equations is set up with 14321 and 15555 data. These equations are solved for each depth. The results are given in Table 2. Because 15555 and 14321 samples differ by a factor of 15 in K contents, the procedure gives the Ar^{39} produced from Ca and from K with errors of the same magnitude as the error in the Ar^{39} measurement. Ar^{39} from K for situation (1) is approximately 15% lower than from situation (2); the Ar^{39} from Ca for situation (1) is 40% higher than from situation (2). With these production rates, the Ar^{39} production rates from K and from Ca for rock 12002 samples and the soil samples are calculated. The sum of the productions from the individual target elements agrees fairly well with the measured Ar^{39} activities for most cases. In soil 10017 the measured Ar^{39} activity is too high. Situation (2) gives a better approximation to the measured activities for the 6 samples that serve as consistency checks than does situation (1). The $K \rightarrow Ar^{39}$ production rate for the top of 14321 is lower than estimated by Begemann *et al.* (1970) for Apollo 11 fines.

The question arises as to how large can the $Fe + Ti \rightarrow Ar^{39}$ rate be made before inconsistencies arise. The Ar^{39} measured activity in sample 15555, 80 limits the $Fe + Ti \rightarrow Ar^{39}$ production rate to less than four times the Reedy and Arnold calculated rates. Therefore, the intensity of solar-flare protons of energy greater than ~ 200 Mev averaged over the past 1000 yr must be less than four times the intensity of the recent galactic cosmic rays if galactic cosmic rays have remained unchanged over 1000 yr.

The next question is whether or not there is any positive evidence from the Ar^{39} measurements for the presence of lower energy protons (< 200 Mev) from solar flares during the past 1000 yr. The depth variation of $Ca \rightarrow Ar^{39}$ does provide positive evidence for low-energy solar flares. The $Ca \rightarrow Ar^{39}$ production rates for all cases decrease slightly with increasing depth from 0 to 5 cm and then increase with increasing depth from 5 to 14 cm. Such a depth dependence is different than that expected for galactic cosmic rays. For example, Reedy and Arnold (1972) calculate that the $Ca \rightarrow Ar^{39}$ from galactic cosmic rays should increase by a factor of 2 in the first 5-cm depth. A direct determination of the intensity of < 200-Mev solar-flare protons averaged over 1000 yr from the $Ca \rightarrow Ar^{39}$ production rates requires knowledge of the Ar^{39} production cross section in thick Ca targets bombarded by protons of less than 200-Mev energy. These cross sections have not been measured. Since neutron-production cross sections for Ar^{39} from K^{39} and from Ca^{40} are known, the neutron fluxes from the $K \rightarrow Ar^{39}$ and $Ca \rightarrow Ar^{39}$ determinations can be obtained and the neutron fluxes can be used to estimate the flux of solar-flare protons.

The $\sigma(n, p)$ cross section in K^{39} decreases from 350 ± 50 mb for 14-Mev neutrons, to 270 ± 40 mb for 4-Mev neutrons, and to approximately 50 mb for 1.5-Mev neutrons (Stehn et al., 1964). Although neutrons of lower energies produce Ar^{39} from K^{39}, we call the neutron flux obtained from the $K^{39} \rightarrow Ar^{39}$ reaction the (> 1 Mev) neutron flux in Table 2, because of energy dependence of the cross section. This flux is 3.7, 10.6, and $12/cm^2$ sec at 1-, 5-, and 12-cm depths, respectively, with a $\sigma(n, p)$ cross section of 200 mb. The (> 1-Mev) neutron flux increases with increasing depth. The $K \rightarrow Ar^{39}$ production rates in 14321 are almost twice the rates calculated by Reedy and Arnold for galactic cosmic-ray production (private communication, 1972).

The $\sigma(n, 2p)$ on Ca^{40} has an 8.1-Mev threshold and is 35 mb for 14-Mev neutrons (Stoenner et al., 1971). The contribution of Ca^{42} to Ar^{39} is small because of its low natural abundance, 0.6%, if the (n, α) Ca^{42} cross section is similar to the (n, α) Ca^{40} cross section. The Ar^{39} from Ca is therefore mainly produced by greater than 10-Mev neutrons on Ca^{40} with approximately a 40-mb cross section. The (> 10 Mev) neutron fluxes obtained from the $Ca \rightarrow Ar^{39}$ column in Table 2 are 2.0, 1.6, and $2.7/cm^2$ sec at approximately 1-, 5-, and 12-cm depths, respectively. The action of galactic cosmic rays on lunar material should produce a neutron flux (> 10 Mev) that increases with increasing depth for the first 14 cm. Evidently, solar flares during the past 1000 yr have increased the > 10-Mev neutron flux in the top centimeter of material by approximately one neutron per cm^2 sec relative to the flux at 5-cm depth. We shall assume that neutrons of greater than 10-Mev energy are produced by solar-flare protons of greater than 50-Mev energy with a cross section of 0.5 b; then 25 protons of (> 50 Mev) energy are required to produce one neutron of (> 10 Mev) energy in the top centimeter. With this estimate, a solar-flare-proton flux (> 50 Mev) of $25/cm^2$ sec or $7 \times 10^8/cm^2$ yr averaged over the past 1000 yr is obtained from Ca → Ar^{39} values in Table 2. On the basis of the Ar^{37} depth dependence after the 24 January 1971 and 2 November 1969 flares, we estimated that approximately $6 \times 10^6/cm^2$ sr or $4 \times 10^7/cm^2$ of protons > 50 Mev were in each of these flares. From the Ar^{39} depth dependence, we estimate that during the past 1000 yr, the flare flux was equivalent to 15 such flares per year.

The measured tritium radioactivities from the samples of 14321 and 15555 and two Apollo 15 soil samples with their approximate depths are given in Table 3. Very little tritium was released at 275°C from the 14321 and 15555 samples. From soil 15271, 17, 15% of the tritium was released at 275°C and from soil 15261, 14, 8% of the tritium was released at 275°C. If the low-temperature tritium observed from the top of rock 12002 (D'Amico <u>et al</u>., 1972) and from soil 15271 is caused by solar-wind implantation of tritium, then the top surface material from our 14321 and 15555 samples, which should contain solar wind, was lost during the sawings and handlings. This is quite likely for 14321, which was a fragile breccia. Rock 15555 is a crystalline rock; our top sample, 15555, 98, was from a broken sloped section (see Figure 2) and could also have had its surface material removed in handling and sawing; however, this is less likely. In both rocks 15555 and 14321, the tritium decreased with increasing depth; however, the decrease was quite small from 6- to 14-cm depth in rock 15555. The decrease of tritium contents with depth is evidence for tritium produced by solar-flare interactions. Figure 5 gives the H^3 depth dependence in the three rocks. The top samples of 15555 and of 14321 had 280 ± 14 dpm/kg compared to 392 ± 11 dpm/kg of tritium in the top of 12002. Differences in the topographies of the rocks probably account for most of tritium difference between the samples at similar depths. We do not consider the differences in tritium contents between samples from 15555, 14321, and 12002 to indicate any serious discrepancies, except possibly for the high tritium in the top sample of 12002. Soil sample 15271, 17 had 300 ± 15 dpm/kg, and soil 15261, 14 had 240 ± 15 dpm/kg of tritium. An estimated 150 dpm/kg of tritium were produced by the action of solar flares in the top centimeter of 14321 and 15555, based on the tritium depth dependences. The solar-flare intensity calculated with this tritium excess is 1.7×10^9 protons (> 30 Mev)/cm^2 yr or 6×10^8 (> 50 Mev)/cm^2 yr with an

Table 3. Tritium activities in Apollo 15 and 14 samples.

Sample	Wgt. (g)	Depth (cm)	Temperature (°C)	H^3 (dpm/kg)	Total H^3 (dpm/kg)	Extraction date
15555, 98	10.65	0−1	275	<3		
		surface	Melt	260 ± 9	281 ± 11	12/10/71
			Remelt	21 ± 5		
15555, 80	10.25	~ 6	275	11 ± 3		
			Melt	146 ± 6	171 ± 8	12/ 2/72
			Remelt	14 ± 3		
15555, 77	8.44	~ 14	275	<3		
			Melt	141 ± 5	166 ± 8	12/23/72
			Remelt	25 ± 5		
14321, 81	10.08	0−0.5	275	<3		
		surface	Melt	270 ± 12	280 ± 14	5/10/71
			Remelt	10 ± 2		
14321, 81	8.49	0.5−1.0	275	5 ± 3		
			Melt	197 ± 6	208 ± 8	5/ 3/71
			Remelt	6 ± 3		
14321, 81	9.46	1.0−1.5	275	6 ± 3		
			Melt	164 ± 8	198 ± 11	4/28/71
			Remelt	28 ± 5		
14321, 267	4.7	~ 5	275	15 ± 5		
			Melt	161 ± 7	176 ± 11	7/20/71
			Remelt	<7		
14321, 95	10.08	~ 12	275	3 ± 3		
			Melt	133 ± 5	145 ± 8	5/25/71
			Remelt	9 ± 4		
Soil 15271, 17	1.97	0−5	275	45 ± 7		
			Melt	255 ± 10	300 ± 15	10/ 7/71
			Remelt	<7		
Soil 15261, 14	2.03	~ 30	275	20 ± 5		
			Melt	220 ± 15	240 ± 15	9/24/71
			Remelt	<4		

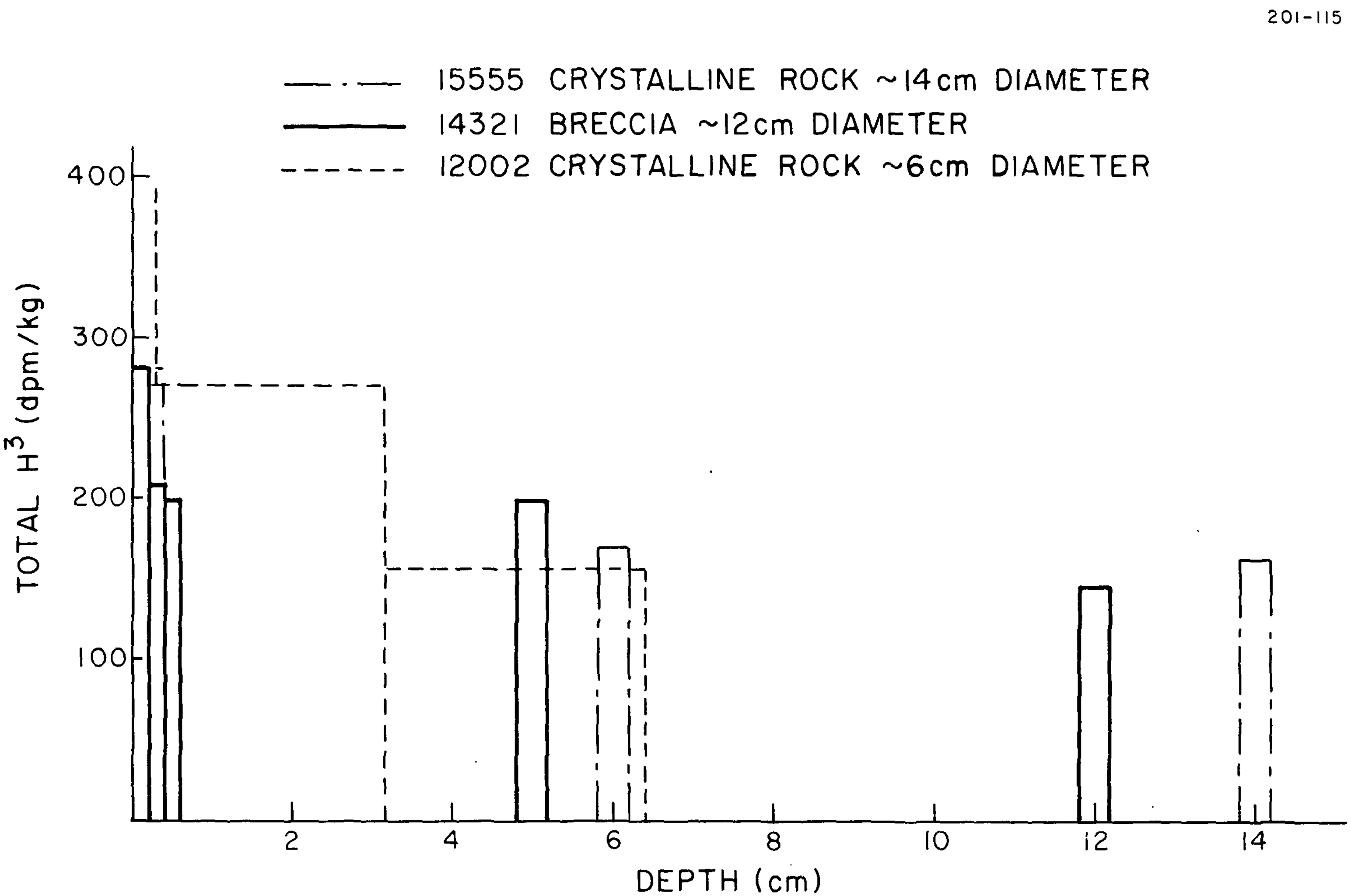

Figure 5. Depth variation of tritium.

E^{-3} differential energy spectrum. The cross sections for this calculation are given by D'Amico et al. (1970). This intensity, which represents an average over the past 30 yr, is nearly the same as that obtained from the Ar^{39}.

The gases from the Apollo 15 soil samples were extracted approximately 2 months after recovery; the gases from rock 15555 samples were extracted 4 to 4-1/2 months after recovery. During most of these times, the samples were stored at room temperature with an atmosphere of either air or dry nitrogen. To examine whether or not this type of storage had any effect on the tritium contents, samples of an Apollo 15 soil, crystalline rock, and breccia were sealed in closed containers with air and hydrogen carrier on 9 August 1971, shortly after the returned sample box was opened. After approximately 3-months' storage at room temperature, the tritium and radon radioactivities in the gas were measured. The results are given in Table 4, where the tritium in the gas of the sealed container is called tritium weepage. No tritium weeped from the crystalline rock sample, which was a sample of 15555. We therefore conclude that the tritium activities measured in 15555 were actually the activities at the time of sample return. The amount tritium weeped from breccia 15565 was only 12.8 ± 1.0 dpm/kg, which is between 5 and 10% of the amounts observed in breccia 14321. If breccia 14321 is similar to breccia 15565 with regard to tritium weepage, then 3 months of storage only reduced its tritium contents by 5 to 10%. However, the tritium weepage from soil 15021, 51 ± 4 dpm/kg, was 20% of the amount observed in soil 15271. If the tritium content of 15271 soil is raised by 20%, its total tritium content is higher than in the top samples from rocks 14321 and 15555 and quite close to the tritium content observed in the top sample of 12002. This may add some support to the suggestion made by D'Amico et al. (1971) that the low-temperature tritium from the top of rock 12002 was solar-wind-implanted tritium.

Table 5 gives the hydrogen and tritium data from Surveyor 3 samples. Although most lunar samples were analyzed with the counter of 42-cm^3 volume and a background of 0.140 ± 0.005 count/min, most Surveyor 3 samples, because of their smaller hydrogen contents, were analyzed with the counter of 7-cm^3 volume and background of 0.0261 ± 0.0014 count/min. The Surveyor samples are very thin, ~ 0.2 g/cm^2 thick. Two of the Surveyor samples were exposed to sunlight while on the moon; these released tritium at 270°C. One of the Surveyor samples was facing the soil and

Table 4. Tritium and hydrogen data from Surveyor 3 samples.

Sample	Wgt (g)	Area (cm^2)	Extraction	Hydrogen (cc STP)	Counter volume (efficiency)	Background (count/min)	Sample (count/min)	H^3 (dpm)	H^3 (dpm/kg)	H^3 (dpm/cm^2)	H^3(sat.) (dpm/kg)
Painted aluminum blank	0.320	1.3	Melt	0.45	42 cm^3 (62%)	0.140 ± 0.005	0.139 ± 0.005	< 0.012			
Painted aluminum blank	0.255	1.0	Melt	0.47	14 cm^3 (55%)	0.048 ± 0.003	0.042 ± 0.003	< 0.008			
Painted aluminum blank	0.335	1.3	20-270°C	0.09*	7 cm^3 (50%)	0.0261 ± 0.0014	0.0269 ± 0.0017	< 0.005			
			270°C-melt	0.30*							
Surveyor 1011.2	0.335	1.3	20-270°C	0.62	7 cm^3 (50%)	0.0261 ± 0.0014	0.0353 ± 0.0016	0.018 ± 0.004	54 ± 12	0.014 ± 0.003	430 ± 95
Some sunlight		-	270°C-melt	0.8.	7 cm^3 (50%)	0.0261 ± 0.0014	0.0375 ± 0.0016	0.022 ± 0.004	66 ± 12	0.017 ± 0.003	530 ± 95
		-	Wall heating and remelt	0.70	7 cm^3 (50%)	0.0261 ± 0.0014	0.0383 ± 0.0016	0.023 ± 0.004	68 ± 12	0.018 ± 0.003	545 ± 95
		-	2nd wall heating and remelt	0.09	7 cm^3 (50%)	0.0261 ± 0.0014	0.0252 ± 0.0016	< 0.004	< 12	< 0.003	< 95
Total								0.063 ± 0.007	188 ± 21	0.049 ± 0.005	1505 ± 170
Surveyor 1002	0.243	1.86	20-270°C	with H_2 carrier	7 cm^3 (50%)	0.0261 ± 0.0014	0.0270 ± 0.0016	< 0.004	< 16	< 0.002	
No sunlight			270°C-melt	with H_2 carrier	7 cm^3 (50%)	0.0261 ± 0.0014	0.0432 ± 0.0016	0.034 ± 0.004	140 ± 16	0.018 ± 0.002	1120 ± 130
			Wall heating and remelt	with H_2 carrier	0.5 cm^3 (68%)	0.0070 ± 0.0010	0.0166 ± 0.0014	0.014 ± 0.002	58 ± 8	0.008 ± 0.001	460 ± 65
			2nd wall heating and remelt	with H_2 carrier	7 cm^3 (50%)	0.0261 ± 0.0014	0.0284 ± 0.0016	< 0.005	< 20	< 0.003	< 150
Total								0.048 ± 0.005	198 ± 25	0.026 ± 0.003	1580 ± 150
Surveyor 1015	0.394	1.67	20-270°C	with H_2 carrier	7 cm^3 (50%)	0.0261 ± 0.0014	0.0651 ± 0.0044	0.078 ± 0.009	198 ± 23	0.047 ± 0.005	1580 ± 185
Most sunlight			270°C-melt	with H_2 carrier	7 cm^3 (50%)	0.0261 ± 0.0014	0.0515 ± 0.0028	0.051 ± 0.006	130 ± 15	0.030 ± 0.004	1040 ± 120
			Remelt	with H_2 carrier	42 cm^3 (62%)	0.140 ± 0.004	0.147 ± 0.004	0.011 ± 0.009	28 ± 23	0.006 ± 0.005	210 ± 185
Total								0.129 ± 0.014	328 ± 36	0.077 ± 0.009	2620 ± 240

protected from sunlight; it released no tritium at 270°C. This sample has 198 ± 25 dpm/kg of tritium. Since 198 ± 25 dpm/kg of the tritium were produced during 2.4 yr, the tritium activity at saturation is 1580 ± 150 dpm/kg. The higher tritium contents in Surveyor 3 material exposed to sunlight than in lunar material indicates that those samples were either contaminated with terrestrial tritium or contain loosely bound tritium that was implanted at the surface by solar wind. Even the Surveyor 3 sample that was not exposed to sunlight had a higher saturation H^3 content than the lunar samples did, which is somewhat of a mystery.

Table 5. Tritium and radon weepage from Apollo 15 samples packaged with air on 9 August 1971.

Sample	Type	Wgt. (g)	Extraction date	H^3 (dpm/kg)	Rn^{222*} (dpm/kg)
15555, 1	crystalline rock	18.7	23 November 1971	<3	–
15565	breccia	9.7	28 October 1971	12.8 ± 1.0	25 ± 5
15021	soil	10.0	2 November 1971	51 ± 4	5 ± 1 2.5 ± 1†

*Counter efficiency for Rn^{222} is estimated to be 2.
†Under vacuum conditions.

The radon emanation was also measured in the sealed containers. The soil under vacuum conditions had a radon emanation rate similar to that observed in the sample container (Stoenner et al., 1971). The samples sealed with air had a higher rate of radon emanation.

5. REFERENCES

BEGEMANN, F., VILCSEK, E., RIEDER, R., BORN, W., and WÄNKE, H.
1970. Cosmic-ray produced radioisotopes in lunar samples from the Sea of Tranquility. Proc. Apollo 11 Lunar Science Conference, Geochim. Cosmochim. Acta Suppl. 1, vol. 2, pp. 995-1007.

BOCHSLER, P., WAHLEN, M., EBERHARDT, P., GEISS, J., and OESCHGER, H.
1971. Tritium measurements of lunar material (fines 10084 and breccia 10046) from Apollo 11, 1971. Proc. Second Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 2, vol. 2, pp. 1803-1812.

D'AMICO, J., DeFELICE, J., and FIREMAN, E. L.
1970. The cosmic-ray and solar flare bombardment of the moon. Proc. Apollo 11 Lunar Science Conference, Geochim. Cosmochim. Acta Suppl. 1, vol. 2, pp. 1029-1036.

D'AMICO, J., DeFELICE, J., FIREMAN, E. L., JONES, C., and SPANNAGEL, G.
1971. Tritium and argon radioactivities and their depth variations in Apollo 12 samples. Proc. Second Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 2, vol. 2, pp. 1825-1839.

FIREMAN, E. L., D'AMICO, J., and DeFELICE, J.
1970. Tritium and argon radioactivities in lunar material. Science, vol. 167, pp. 566-568.

REEDY, R. C., and ARNOLD, J. R.
1972. Interaction of solar and galactic cosmic-ray particles with the moon. Journ. Geophys. Res., vol. 77, pp. 537-555.

STEHN, J. R., GOLDBERG, M. D., MAGURNO, B. A., and WIENER-CHASMAN, R.
1964. Neutron cross sections. Brookhaven Nat. Lab. Rep. 325, 2nd ed., Suppl. No. 2.

STOENNER, R. W., LYMAN, W. J., and DAVIS, R., Jr.
1970a. Cosmic-ray production of rare gas radioactivities and tritium in lunar material. Science, vol. 167, pp. 553-555.

1970b. Cosmic-ray production of rare gas radioactivities and tritium in lunar material. Proc. Apollo 11 Lunar Science Conference, Geochim. Cosmochim. Acta Suppl. 1, vol. 2, pp. 1029-1036.

STOENNER, R. W., LYMAN, W. J., and DAVIS, R., Jr.

1971. Radioactive rare gases in lunar rocks and in the lunar atmosphere. Proc. Second Lunar Science Conference, Geochim. Cosmochim. Acta, Suppl. 2, vol. 2, pp. 1813-1823.

WAHLEN, M., HONDA, M., FRUCHTER, J., FINKEL, R., KOHL, C., ARNOLD, J., and REEDY, R.

1972. Cosmogenic nuclides in football-sized lunar rocks. Abstracts of Third Lunar Sci. Conf., pp. 764-766.