

three weak exotherms: (A) 250° to 450°C, < 4 cal/g; (B) 450° to 710°C, < 20 cal/g; and (C) 750° to 850°C, 13.5 cal/g. Peaks A and B are suppressed when heated in N₂. Peak A varied from <0.5 cal/g to ~5.7 cal/g in different core samples. Peak C is relatively sharp and has two subpeaks. Rock 57 shows no DTA peaks. The absence of peak C in rock 57 suggests that peak C may be associated with the glass in the fines. This would be consistent with two other observations: (i) at ~800°C, several euhedrally shaped glassy grains were observed by us to burst explosively; and (ii) x-rays showed reversion of a glassy spherule to a feldspathic crystalline material after heating at 900°C. From the DTA data we conclude that there is no large stored energy release due to the recombination of lattice defects produced by the lunar radiation environment.

Perhaps our most interesting result is the observation of solar flare effects. From studies of tracks in internal grains from breccias and in the surfaces of crystalline rocks that have been buried at different times in the past, lunar samples can be used to study the history of solar activity.

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8. This work would have been impossible without the assistance of P. Swan. We also wish to thank M. Marberry for her invaluable help. Much of the laboratory work was done by A. Jones. This work was supported in part by contracts from NASA and the McDonnell Douglas Corporation. Dr. Crozaz is Chargé de Recherches du Fonds National Belge de la Recherche Scientifique, on leave; Dr. Haack is on leave from University of Göttingen, Germany; Dr. Maurette is on leave from Institut de Physique Nucléaire, Laboratoire de Spectrométrie de Masse, Orsay, France; Dr. Miyajima is on leave from Waseda University, Japan; and Dr. Wittels is on leave from the U.S. Atomic Energy Commission.

4 January 1970

Tritium and Argon Radioactivities in Lunar Material

Abstract. Tritium and argon radioactivities, attributable to galactic and solar cosmic-ray interactions, were measured in lunar soil and in three lunar rocks. The tritium in the soil, 325 ± 17 disintegrations per minute per kilogram, is slightly higher than that in the rocks, 212 to 250 dpm/kg. For two rocks, the tritium was combined with the helium-3 in order to calculate exposure ages of 375 ± 40 and 205 ± 25 million years. The argon-37 radioactivities, 21.0 to 27.2 dpm/kg, and the argon-39 radioactivities, 12.1 to 16.4 dpm/kg, are slightly higher than those in stony meteorites. Higher exposure ages were obtained from the argon isotopes than from tritium and helium-3. On the basis of the known galactic cosmic-ray flux and the known cross section, at least half of the observed radioactivities are produced by solar cosmic rays.

The amounts of tritium and argon radioactivities in lunar material are of interest because they give information about galactic and solar-source mechanisms and because they help determine exposure ages. These ages are related to the erosion history of the lunar surface.

The apparatus used for the argon measurements in lunar materials is similar to that for argon measurements in meteorites except for the improved proportional counters, whose characteristics are given in a recent article (1). For tritium, counter and other improvements made determinations possible in samples as small as 1 g.

Lunar rocks 72, 17, and 61, of petrological types A, B, and C, respectively, and a sample of soil fines, which have been described by the Lunar Sample Examination Team (2), were analyzed. The samples were melted under vacuum in the presence of argon carrier at 1600°C for 2 hours. The evolved gases were reacted over vanadium foil at approximately 800°C. The noble gases were removed from the vanadium foil at room temperature, and their volumes were measured. The helium and neon were separated by freezing the argon and heavier noble gases on charcoal at liquid-nitrogen temperature. Carrier krypton was added to the charcoal. The argon was separated from the krypton at Dry-Ice temperature and placed in low-level proportional counters with volumes between 0.54 and 0.75 cm³.

The argon pressure in the counters was standardized at 1.4 atm. The argon yields were between 95 and 99 percent. Methane (10 percent) was added to the argon. The counters were removed from the extraction system and counted in a low-level counting system where the backgrounds were 1 count/day or less in 2.8 ± 0.6 kev channels of a 100-channel analyzer. The 2.2 to 3.4 kev energies cover the ³⁷Ar peak to 1/10 maximum. The backgrounds were less than 4 count/day above the 4.0-kev channel where the ³⁹Ar was counted. The counting efficiency for ³⁷Ar in different counters varied from 41 to 45 percent; and for ³⁹Ar, from 14 to 20 percent. A thin quartz window at one end of the counter permits the passage of the 5.9 kev x-ray from an ⁵⁵Fe source used to adjust the energy scale. Calibration curves with known amounts of ³⁷Ar and ³⁹Ar for a typical counter are given in another publication (1). Figures 1 and 2 show the argon counting data, together with backgrounds, from the 9.9-g lunar soil sample for 9061 minutes between 16 and 23 September 1969, and from the 10.0-g sample of lunar rock 17 for 12,936 minutes between 3 and 13 October 1969. The argon samples were counted for approximately 2 months, and the ³⁷Ar activity decayed with a 35-day half-life.

The krypton was removed from the charcoal at 0°C and put into an argon-type counter to which methane had been

Table 1. Radioactivities and gas contents from lunar samples. The only krypton data obtained were from soil 84-24 (dpm/kg): ⁸¹Kr, 0.14 ± 0.10 ; ⁸⁶Kr, < 1.

Sample	Weight (g)	Tritium (dpm/kg)	Hydrogen (cm ³ /g)	Helium (cm ³ /g)	³⁹ Ar (dpm/kg)	³⁷ Ar* (dpm/kg)
<i>Apollo 11</i>						
Soil 84-24	(9.9)	325 ± 17	1.2	0.26	12.1 ± 0.7	27.2 ± 2.2
Rock 17-14	(10.0)	219 ± 7	0.47	$< 5 \times 10^{-3}$	16.4 ± 0.9	21.0 ± 2.0
Rock 72-11	(9.4)	234 ± 10	0.76	$< 5 \times 10^{-3}$	15.8 ± 1.0	25.7 ± 2.0
Rock 61 surface	(1.49)	235 ± 15	2.5	~ 0.3		
Rock 61 interior	(0.96)	231 ± 10	1.4	~ 0.4		
<i>Meteorite(1)</i>						
Sprucefield L4 (nonmagnetics)	(21)	290 ± 30		$< 5 \times 10^{-3}$	7.7 ± 0.4	12.6 ± 1.4

*Extrapolated to 21 July 1969.

added. The electronics were adjusted so that the 5.9 keV x-ray from the ^{55}Fe source was peaked in channel 30 of the 100-channel analyzer. The 11.9 keV peak from ^{81}Kr should then appear in channel 61. The counter efficiency for ^{81}Kr was assumed to be the same as for ^{37}Ar , and for ^{85}Kr , to be 30 percent above 4.0 keV.

The hydrogen was removed from the vanadium foil while heated, and the volume of the released hydrogen was measured. The hydrogen counter consisted of a copper cathode and a 0.002-inch Therlo center wire in a quartz envelope that had a thin window at one end; its total volume was 42 cm³. The electronics were adjusted so that the 100-channel analyzer scanned energies up to 19.3 keV. With 400-torr pressure of P-10 gas (commercially available counting gas containing 90 percent argon and 10 percent methane) and 200-torr pressure of hydrogen, the counting efficiency for tritium between 1 and 19.3 keV was 60 percent. The resolution for the 5.9 keV source was 20 percent. The background was 0.14 count/min. The tritium spectrum does not consist of a sharp peak; however, it has a different shape from the background spectrum.

Table 1 summarizes the results. For comparison, the radioactivities in the nonmagnetics of the recently fallen Sprucefield meteorite, an L4 chondrite, were measured with the same equipment. The radioactivities in the lunar material are within a factor of 2 of those in the meteorite. There are, however, interesting differences between the lunar samples and also between lunar samples and meteorites.

The tritium in the soil was 325 ± 17 dpm/kg; in rocks 17 and 72, it was 219 ± 7 and 234 ± 10 dpm/kg. A chip from the surface of rock 61, which on the basis of pit count is thought to be the top side, had 235 ± 15 dpm/kg; an interior sample of the same rock had 231 ± 10 dpm/kg. The higher content in the soil cannot be caused by the small differences in chemical composition between the soil and rocks. The helium content of the soil was 0.26 cm³/g, in accord with the mass spectrometer results (2); this content was attributed to the solar wind. The hydrogen in the soil was 1.2 cm³/g, which may in part be solar-wind hydrogen. However, the difference between the tritium in the soil and that in the rocks should not be attributed to solar-wind tritium until other possibilities are eliminated. On the basis of the small difference in the tritium contents of the surface and interior sam-

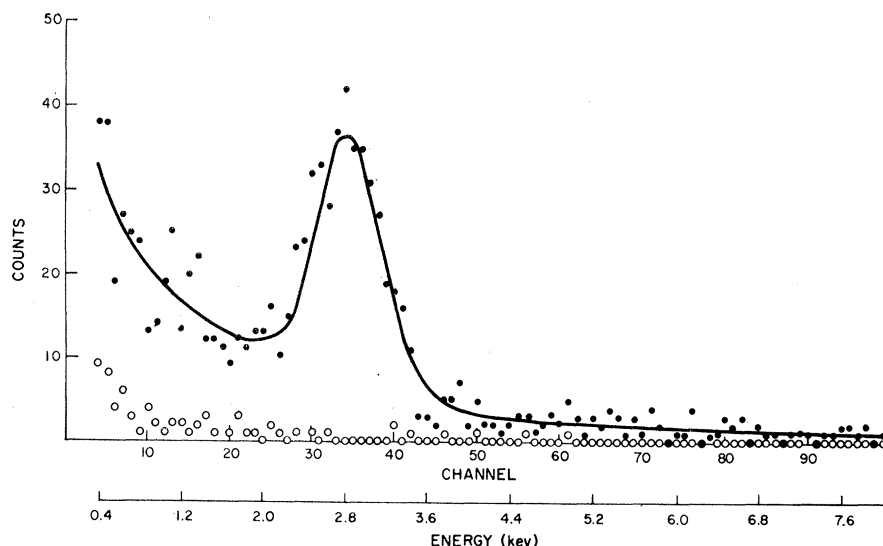


Fig. 1. Argon radioactivity for 9.9 g of lunar soil. Solid line, curve from calibrations with $^{39}\text{Ar} = 12.1$ dpm/kg and $(^{37}\text{Ar})_0 = 27.2$ dpm/kg. Solid circles, counts per channel for 9061 minutes 57 to 64 days after 21 July 1969 (there were 123 counts beyond channel 99). Open circles, background counts per channel for 11,426 minutes (there were 11 counts beyond channel 99).

ple of rock 61, one could set an upper limit of 0.5 triton/cm² sec for tritium implanted by solar wind, if the surface sample was the top side. However, tritium from solar flares, which would not be implanted at the surface, might be the source of the difference. There is a possible difference in the average depths of the soil and rock material that could account for the tritium difference. More detailed studies of tritium versus depth are necessary before the excess tritium in the soil can be interpreted.

The tritium contents of rocks 17 and 72 can be combined with their spallation ^3He contents of $(3.2 \pm 0.2) \times 10^{-6}$ and $(1.9 \pm 0.1) \times 10^{-6}$ cm³/g meas-

ured at the Lunar Receiving Laboratory (3) to obtain cosmic-ray exposure ages. The exposure age of rock 17 is 375 ± 40 million years, and that of rock 72 is 205 ± 25 million years, with the ^3He production rate assumed to be twice the present tritium decay rate. These ages are simply interpreted as the time since the rocks have been thrown out from large impact craters. There are two possible complications to this simple interpretation: some ^3He may be lost by diffusion, and the production of ^3He may not have been constant because of changes in the amount of surrounding material with time. The rocks have been eroded by small-particle bombardment,

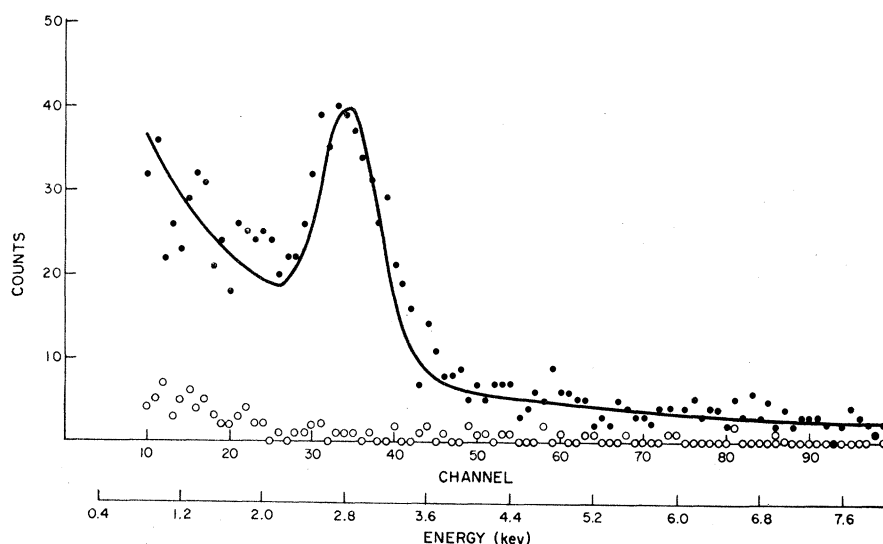


Fig. 2. Argon radioactivity for 10.0 g of lunar rock 17. Solid line, curve from calibrations with $^{39}\text{Ar} = 16.4$ dpm/kg and $(^{37}\text{Ar})_0 = 21.0$ dpm/kg. Solid circles, counts per channel for 12,936 minutes 74 to 84 days after 21 July 1969 (there were 178 counts beyond channel 99). Open circles, background counts per channel for 13,078 minutes (there were 22 counts beyond channel 99).

as indicated by minute pits on the rounded surfaces (2). The soil may have been in a state of flux because of this bombardment and because of internal stresses. Consequently, the rocks could have been buried in the soil at various times in the past. It is therefore of interest to obtain exposure ages from the argon isotopes.

The ^{39}Ar activity in the soil was 12.1 ± 0.7 dpm/kg, and those in rocks 17 and 72 were 16.4 ± 0.9 and 15.8 ± 1.0 dpm/kg. The ^{37}Ar activity in the soil was 27.2 ± 2.2 dpm/kg, and those in rocks 17 and 72 were 21.0 ± 2.1 and 25.7 ± 2.0 dpm/kg. An exposure age could be obtained from combination of spallation ^{38}Ar and the argon radioactivities if the relative production rates were known. To a first approximation, the ^{38}Ar production rate equals that of ^{37}Ar plus ^{39}Ar . The ^{38}Ar produced by cosmic rays in rocks 17 and 72, as measured at the Lunar Receiving Laboratory (3) is $(50 \pm 10) \times 10^{-8}$ and $(33 \pm 5) \times 10^{-8}$ cm³/g, respectively. The ^{38}Ar to the ^{37}Ar plus ^{39}Ar exposure age of rock 17 is 640 ± 160 million years, and that of rock 72 is 410 ± 80 million years. These ages are 30 percent or more higher than the ^3He to ^3H ages and indicate either that some ^3He has been lost or that the amounts of material surrounding the rocks have changed during the past 400 million years.

The tritium and argon radioactivities in meteorites are attributed to the interactions of galactic and solar cosmic rays with the material. It is reasonable to attribute these radioactivities in lunar material to the same mechanism. Since the flux of galactic cosmic rays at 1 A.U. is known (4) and the depth of the lunar samples is fairly small (less than 25 g/cm²), one can estimate what fraction of the radioactivities are attributable to galactic cosmic rays. The flux of nucleons with energy greater than 1000 Mev is 0.8 ± 0.1 nucleon/cm² sec and that with energies between 400 and 1000 Mev is 0.35 ± 0.1 nucleon/cm² sec averaged over the solar cycle. The total tritium-production cross section in oxygen is 38 millibarns, independent of the bombardment energy above 400 Mev (5). For heavier elements, the tritium cross section increases with atomic weight A as approximately $A^{2/3}$ for particles above 1000 Mev and is roughly independent of atomic weight for particles between 400 and 1000 Mev (6). With these fluxes and cross sections, we obtain a dpm/kg, which is less than half the tritium production rate of 85 ± 15

amount observed. Similar considerations for the ^{39}Ar production from iron and titanium lead to the conclusion that only one-quarter or less of the ^{39}Ar observed in the lunar rocks is produced by galactic cosmic rays. It therefore appears that at least half the observed radioactivities are produced by solar cosmic rays.

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Particle Track, X-ray, Thermal, and Mass Spectrometric Studies of Lunar Material

Abstract. *Particle tracks in Apollo 11 samples are dominantly of cosmic ray and solar origin: primary galactic and solar flare particles, likely spallation recoil tracks, and possible solar-wind heavy particles. The energy spectrum of iron-group nuclei is inferred from track density gradients in surface layers, and a limit of $< 10^{-7}$ centimeter per year is deduced for the surface erosion rate. From cosmic ray tracks in rock and core samples it is clear that the lunar soil is stirred often during each few million years. X-rays reveal augite, anorthite, olivine, ilmenite, troilite, nonmeteoritic iron, and assorted glasses, but no major structural damage. Hydrogen, helium, and other gases in the fines are compatible with expected solar wind ratios.*

In addition to being of direct crystallographic and thermodynamic interest, phase identification is vital to proceeding with particle track studies. X-ray, electron microprobe, and optical, chemical etching, and standard mineral separation techniques were used. Debye-Scherrer patterns revealed that the dominant crystalline phases in the lunar fines (sample 2,84) are pyroxene and feldspar. Selected (diameter, > 50 μm) crystals from the coarse fines (sample 2,85) and from the interior of rock 44 (sample 44,47) were examined by Laue transmission and x-ray precession camera techniques. They revealed anorthite, pyroxene, olivine of $30 (\pm 5)$ percent fayalite, and ilmenite. The crystals were generally of good quality without major structural defects. Twinning occurs in some larger crystals, an example of deformation twinning in ilmenite being visible in Fig. 1. Microprobe studies of this sample (rock 2,85,4) revealed the same phases plus troilite and free iron (nonmeteoritic, since nickel was not detectable). The pyroxene was determined to be augite. Specific crystals identified by the microprobe are indicated in Fig. 1. Comparison of this section with a thin section in both transmitted and reflected light provided us with a means of rapid visual identification of most of the crystalline phases of interest for track

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5 January 1970

etching (1). Glass was readily identifiable by its response to etching of particle tracks in hydrofluoric acid (2).

Our objectives are, first, to identify the variety of particle tracks present in lunar materials; second, to use these tracks to learn about the history of the moon; and finally, to learn about the solar and galactic cosmic rays that impinge upon the moon. Possible track origins in extraterrestrial materials have been described (3, 4), and examples have been given of the identification of several varieties in meteorites (5-7). In this work we report the observation of tracks thought to be solar flare particles, spallation recoil tracks in glass, and possible solar-wind heavy nuclei in a crystalline phase.

The most abundant tracks found in this study are from heavy iron-group nuclei (Fig. 2a). They have been identified by criteria we have noted elsewhere (3, 5): track densities decreasing toward the interior (rocks 3,44, and 17); preferred orientation of tracks (rocks 3,44, and 5-2.6,1,3); approximately equal track densities in adjacent crystals of different mineral types (rocks 2-85,4; 3; 17; and 44); and length distribution peaked at ~ 10 μm , but including shorter and longer tracks (rocks 3,44, and 5,2.6,1,3) and present in crystals (anorthite and augite) having too little uranium ($\leq 10^{-10}$ weight frac-