The lunar crystalline rocks show a wide isotopic variation in krypton and xenon, all characterized by excesses of xenon 124–132 and krypton 78–84 relative to ^{136}Xe and ^{86}Kr, respectively. These excesses are attributed to cosmic-ray spallation reactions in the rocks, mainly on Ba, Sr, Y, and Zr. The spallation spectrum for the various rocks are all similar and are (normalized to $^{126}Xe \equiv 1$ and $^{83}Kr \equiv 1$): 124/126/128/129/130/131/132/134 $\equiv$ 0.55 $\equiv$ 1/1.5/1.7/1.07/6.3/0.9/0.07, and 78/80/82/83/84 $\equiv$ 0.2/0.48/0.76 $\equiv$ 1/0.38. These spectra are quite similar to the spallation spectra found in achondrites except for a much higher ^{131}Xe and a somewhat lower ^{84}Kr. The higher 131 yield is probably an effect of low energy protons as barium is the major target for spallation Xe production, and 730-mev proton spallation of barium does not show this high ^{131}Xe yield (*5*). The amounts of ^{126}Xe and ^{83}Kr in the rocks vary by almost an order of magnitude among samples because of the differing exposure histories, yet these amounts correlate reasonably well with the spallation ^{38}Ar. The slight isotopic excesses of the lighter isotopes of Kr and Xe in the lunar fines and breccia over that in carbonaceous chondrites is also probably due to spallation reactions. Not only is the amount of excess ^{126}Xe in different breccia variable, but in amount it is only slightly greater than the spallation ^{126}Xe found in those crystalline rocks with the longest exposure history.

Radiogenic Ar and He are apparent in all crystalline rocks examined. K-Ar ages were obtained for 14 rocks and fragments for which K was known (*1*). Potassium was determined by emission spectroscopy but not from aliquots used for noble gas analysis. The K-Ar ages, including two from lithic fragments from two breccia samples, range from 2.5 to 3.8 billion years (Fig. 1). The U-He ages are subject to large errors because of the solar wind correction to the measured ^{4}He and the uncertainty in the U-Th content derived from the K values (*1*); nevertheless, most of the ages are concordant within this error with the K-Ar data. Thus, the spread in the K-Ar ages is probably real to some extent, and not just caused by variable leakage of radiogenic Ar.

Mare Tranquillitatis, then, probably first crystallized about 4 billion years ago from a large molten body produced either by meteoritic impact or volcanism. Subsequent large impacts or volcanic events would further mold the mare to its present form and produce younger rocks. Despite the time spread of approximately 1 billion years in volcanic events, it appears that the features of the moon at Mare Tranquillitatis were well delineated at least 2 billion years ago.

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Solid State Studies of the Radiation History of Lunar Samples

Abstract. *Particle track densities up to $> 3 \times 10^9$ per square centimeter have been measured in different samples. Rocks 17, 47, 57, and 58 have VH ($Z > 22$) galactic cosmic ray ages of 11, 14, 28, and 13 $\times 10^6$ years, respectively. Rock 57 has a calculated erosion rate of $\lesssim 10^{-7}$ centimeter per year. Near-surface track versus depth data in rock 17 can be fit with solar flare particles that have a differential energy spectrum αE^{-3}; lunar samples can be used to study the history of solar activity. The uranium in the crystalline rocks occurs principally in small regions <10 to ~100 micrometers in size. The (low) thermoluminescence of the fines increases with depth in core 10004. With one possible exception, x-ray studies have not shown pronounced radiation damage effects. The total energy release upon heating is small up to 900°C and occurs in three broad regions.*

We have measured particle tracks, thermoluminescence, x-ray distortions, U distributions, and energy release patterns in lunar samples. Table 1 gives a summary of track data. The lower track densities were measured with an optical microscope and the higher densities with a Cambridge Mark IV stereo-scan electron microscope. A series of cross calibrations (Fig. 1) have convinced us that the geometrically shaped, deep surface holes normally counted in the stereo-scan are essentially equivalent to the long tracks seen in the optical microscope after etching.

Track densities versus depth were measured in rocks 17 and 57. For the following reasons we believe that the tracks found at $\gtrsim 5$ mm from an external surface are due primarily to VH nuclei ($Z > 22$) from galactic cosmic rays: (i) the uranium concentrations are too low to give an appreciable spontaneous fission track density; (ii) the tracks are frequently anisotropically distributed and have a length distribution characteristic of VH tracks studied in meteorites; (iii) long tracks attributable to VVH nuclei ($z \gtrsim 31$) are observed with an abundance of $\sim 10^{-3}$ of the total density; and (iv) deeper than 5 mm, there is an accord between theory and experiment for the depth profile of galactic tracks.

Using feldspar data from interior portions we calculate the following VH exposure ages: (i) 10017, $11 \pm 1 \times 10^6$ years, (ii) 10047, $14 \pm 4 \times 10^6$ years, (iii) 10057, $28 \pm 5 \times 10^6$ years, (iv) 10058, $13 \pm 2 \times 10^6$ years. The quoted errors are statistical. Greater uncertainties arise from the lack of precise sample position in the rocks. Our calculations are based on previous theoretical treatments (*1*) using a modified low energy spectrum (*2*). Because all the surfaces, including "tops" and "bottoms," have high densities compared with densities of interior samples, we use a "rolling stone model" in which the irradiation is uniform on all sides.

The VH exposure ages are all approximately equal and are in striking contrast to the proton spallation ages that can be inferred from the rare gas data reported in the preliminary examination. From both ^{3}He and ^{21}Ne data, rock 17 has the oldest spallation age, probably $\gtrsim 200 \times 10^6$ years. Rock 57, in contrast, has the youngest spallation age, probably $\gtrsim 40 \times 10^6$ years.

The general similarity of the VH ages could be the result of an equilibrium process such as erosion or thermal annealing of tracks. However, the considerable difference in the ages of rocks 17 and 57 argues against this view.

The simplest interpretation of the low ratio of VH to spallation ages in rocks 17, 47, and 58 is to assume that these rocks were buried at least 15 cm (17) and 11 cm (47, 58) below the surface during most of their spallation age. If this explanation is correct, we would expect more isotope production by secondary nuclear cascade products in these rocks than in rock 57.

With the method outlined by Price *et al.* (*3*), we found that the accord between the VH and spallation ages in rock 57 can be used to set an upper limit of $\lesssim 0.1$ cm/10^6 years for the surface erosion rate of this rock. This value is in accord with the surface track data discussed next.

Starting at about 5-mm depth, the track density rises abruptly as the surface is approached, reaching values $>3 \times 10^9$ per square centimeter. The VVH/VH ratio is $\sim 10^{-3}$. The densities are highly variable from one surface grain to the next and frequently show strong gradients within a single grain. The profiles of density versus depth in the first 30 μm of certain grains are quantitatively similar to the profiles reported by Pellas *et al.* and by Lal and Rajan (*4*) for grains in the meteorite Kapoeta. The extrapolated galactic spectrum can account for <5 percent of these surface tracks, and we attribute them to either solar flares (most likely) or to a demodulated galactic spectrum (less likely) very different from that currently observed in periods of low solar activity.

We have fitted out 0- to 5-mm depth data using the following differential energy spectra (*5*): $S_1 = 8 \times 10^{12} E^{-3}$ p/cm^2 10^6 years sr/Mev/amu, $S_2 = 1.6 \times 10^{11} E^{-2}$, and $S_3 = 1.9 \times 10^{14} E^{-4}$. These spectra are normalized to give an integral VH flux of 10^5 p/cm^2 year for an energy ≥ 20 Mev/amu, averaged over a solar cycle. Within broad errors due to sampling difficulties, the results in rock 17 can be fit with the spectrum S_1 between 1 and 5 mm. Below 1 mm this spectrum predicts higher densities than are observed and does not agree with the gradients measured in surface grains. However, agreement can be restored by taking an erosion rate of slightly less than 0.1 cm/10^6 years and by assuming that the rock was lightly covered with a thin layer of dust. The spectrum S_3 definitely does not fit the data.

Most crystals removed from the <1-mm fraction of the fines and from the breccias show high track densities similar to those observed in surface crystals from rocks. Some also show edge effects and the tracks are probably produced primarily by solar flares. If rock erosion proceeds gently by the flaking off of individual surface grains, which have already received a solar flare irradiation, then we would conclude from the track densities observed in the fines that crystals are removed (and subsequently covered over) at the rate of $\sim 2 \times 10^{-7}$ cm/year or one layer of 100 μm crystals every 5×10^4 years. The core results suggest a possible depth dependence from which average turnover rates could be calculated; however, more data are needed. The large amount of rare gas in the fines cannot be accounted for by stopping flare particles and must be due to the solar wind.

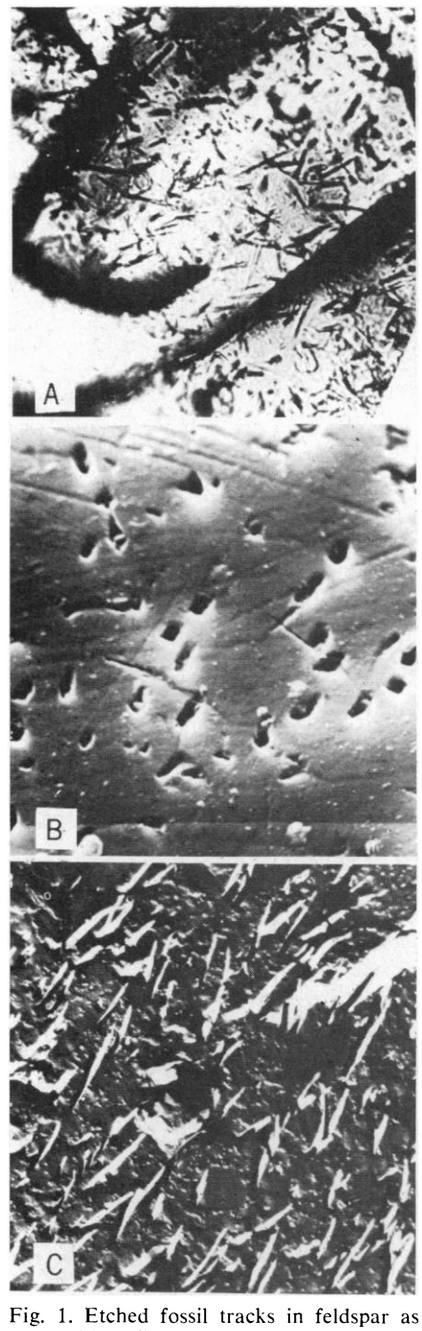

Fig. 1. Etched fossil tracks in feldspar as seen with different types of instruments: (A) in the optical microscope, tracks appear as long dark lines; (B) in the stereoscan electron microscope, the tracks show as deep, geometrically shaped holes; (C) plastic replicas viewed with a transmission electron microscope prove that the holes seen in the stereo-scan at high track densities are not superficial surface pits. Agreement is found between these three methods when measurements are made on the same crystal (not shown here).

Track densities measured in the internal portions of glass spherules are highly variable, with ~80 percent of the spherules having densities too low to measure. Tracks are difficult to observe on the external surfaces, which are frequently covered with holes of a quite different origin as well as secondary glass splatters, impact pits, and a variety of other interesting features. The differences in track densities from one spherule to the next are probably due both to different radiation histories and to different track retentivities. Track annealing temperatures (1 hour) in spherules of different composition range from ~100°C to >400°C, suggesting the use of spherules as temperature monitors. Tracks that contain spherules frequently have an edge effect similar in profile to that observed in surface grains. The edge effect is generally uniform, which indicates an isotropic irradiation. In some spherules the track pits fall into two size classes, with the larger resembling fission tracks. If this were proved to be the case, some spherules would have ages of $\sim 10^9$ years. Dating of the formation time of some spherules is likely to prove possible, particularly in the case of nonhomogenized spherules which contain small inclusions with high uranium concentrations.

Uranium maps of individual grains and rock fragments were made by registering thermal neutron-induced fission tracks either in external mica detectors or in the annealed grains themselves (*6*). In all the materials, with the exception of some glass spherules that have uniform distributions of U in the range 150 to 500 ppb, the U occurs in small regions ranging in size from <10 to ~ 100 μm and with concentrations from 1 to ~ 100 ppm. About one-

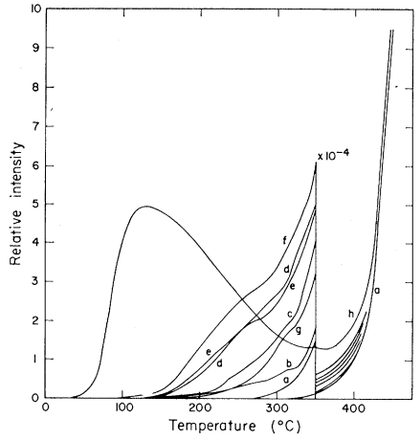

Fig. 2. Thermoluminescence (TL) glow curves in unseparated lunar fines from core 10004. Light output is measured as a function of temperature in samples heated rapidly in argon gas. The sample is then cooled, irradiated with ^{90}Sr β rays, and reheated to calibrate its response. The TL output increases with increasing depth. The high temperature β-ray response is the same for all these samples. (a) background; (b) core tube, top; (c) core tube, 3 cm from top; (d) core tube, 6 cm from top; (e) core tube, 9 cm from top; (f) core tube, 12 cm from top; (g) lunar fines; (h) core tube, top, 1 hour.

third of these regions are associated with discrete objects that range in color from clear white to black. Apart from inclusions, the average U concentrations in feldspars, pyroxenes, and ilmenites were measured as <50 ppb by one method and, in fewer samples, as <5 ppb by a second, more sensitive method. Powder samples of the bulk fines and of the breccias 46 and 23 had uranium concentrations of 550, 610, and 620 ppb, respectively. We conclude that the uranium resides principally in several different small grain phases. These phases, which are likely linked to trace elements such as Zr and P, would have uranium concentrations high enough to date by the fission track method.

The x-ray measurements were made with Debye-Scherrer, Laue, and precession camera techniques to identify minerals and to look for crystal distortions characteristic of radiation damage. In addition to the major minerals, we found a total of six red grains ranging in size from 20 to 200 μm in samples from core 10004. Two small grains were found in the surface sample, three found at 6 cm deep, and another at 9 cm. From their space group and lattice spacings, they were identified as alpha aluminum oxide. Most of the grains from the fines and breccias showed no obvious crystalline distortion. In some crystals the Laue spots formed continuous streaks similar to those we have previously observed in samples of feldspar and pyroxene irradiated with heavy ions (*7*). However, contrary to the minerals irradiated with heavy ions, these distortions did not anneal at temperatures of 900°C. Lattice parameter measurements were made on the fine material before and after annealing at 900°C. These measurements showed less than a 10^{-3} fractional change in spacings for both pyroxenes and feldspars above 200-mesh in size. This is consistent with what we would predict from our track studies on lunar materials. Pyroxenes below 400-mesh in size, where the effects of solar wind and solar flare particles should be most easily detected, show a change in spacing of up to 0.5 percent upon annealing. This effect may be due to radiation, although other explanations are possible.

Thermoluminescence (TL) was measured in an argon atmosphere to 500°C with an EMI9635QB photomultiplier. The natural TL is low and is not saturated in any sample in spite of the large radiation doses on the moon.

The TL output of lunar fines increases with depth in core 10004 (Fig. 2). From these data we infer that the average depth of bulk lunar fines was ~2 cm. The natural and β-ray sensitivities of the oriented breccia 46 are similar to the fines, but no clear-cut depth effect is seen.

Both fines and breccias show a large TL peak at ~ 120°C when irradiated with ^{90}Sr β rays. Partial annealing of β-irradiated samples gives curves similar to the natural TL curves. Heating of a sample to >800°C for 8 hours fails to modify the subsequent β response. A low temperature peak is also present in a surface sample of rock 57, but it is missing in an interior sample. It is also absent in both external and internal samples of rock 47. The ratio of natural TL to β-ray TL was ~5 times greater at the surface than in the interior of rock 57 but not in rock 47.

Handpicked grains from rock 47 show that its TL output is due primarily to feldspar. The results indicate that separation of feldspars should greatly enhance the TL signal in the fines.

Since the ionization does not increase with depth, it is tempting to associate the core results with the temperature gradient in the lunar surface. Detailed kinetic studies, as well as low-temperature irradiations and dose rate studies, are in progress to test this hypothesis.

Measurements of total energy release were made with a Dupont model 900 differential analyzer. When heated in air to 900°C, the fines and breccias give

Table 1. Summary of fossil track results. These results do not include the surface crystals which have densities that range above 3×10^9 tracks per square centimeter and which depend critically on the position with respect to the surface.

Sample	Density range (tracks/cm²)	Average density (tracks/cm²)
	Crystalline rocks	
10047 int. ~5 mm from surface	5×10^6 to 1.7×10^7 (12 feldspars)	$(1.2 \pm 0.4) \times 10^7$
	4×10^6 to 8.4×10^6 (13 pyroxenes)	$(5.6 \pm 0.4) \times 10^6$
10058 int. ~12mm from surface	7 to 7.7×10^6 (2 feldspars)	$(7.4 \pm 0.6) \times 10^6$
	4.4 to 6.4×10^6 (8 pyroxenes)	$(5.6 \pm 0.3) \times 10^6$
10057	12 positions in vertical section give monotonic variation from 1.5×10^7 at center to $> 10^9$ at surfaces	
10017	12 positions in vertical section give monotonic variation from 3.0×10^6 at center to $> 10^9$ at surfaces	
	Breccia	
10046 ~15mm from surface	$\sim 10^9$ (2 feldspars); many others with reaction layer; out of 5 dark spherules, one with $\sim 2 \times 10^7$ (small pits), another with $\sim 5 \times 10^5$ (large pits)	
	Fines	
Olivines (5)	1.6×10^7 to 2.3×10^7	$(1.8 \pm 0.2 \times 10^7)$
Feldspars (30)	1.5×10^7 to $> 10^9$	
Pyroxenes (30)	6×10^6 to $> 5 \times 10^8$	
Spherules (36)	Six dark spherules with densities that ranged from 2×10^7 to 1.2×10^8 (small pits); three of them with $\sim 10^5$ (large pits); other spherules contain only shallow pits.	
	Coarse fraction of fines	
Pyroxenes from 2-mm fragment (2)		$\sim 5 \times 10^6$
	Core tube (4 crystals for each location)	
Surface	10^9 to 2×10^9	
6 cm below surface	6×10^8 to 10^9	
12 cm below surface	2.6×10^7 to 3.5×10^8	

three weak exotherms: (A) 250° to 450°C, < 4 cal/g; (B) 450° to 710°C, $\lesssim$ 20 cal/g; and (C) 750° to 850°C, $\sim$13.5 cal/g. Peaks A and B are suppressed when heated in N_2. 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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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 cm^3. 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 ^{37}Ar peak to 1/10 maximum. The backgrounds were less than 4 count/day above the 4.0-kev channel where the ^{39}Ar was counted. The counting efficiency for ^{37}Ar in different counters varied from 41 to 45 percent; and for ^{39}Ar, 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 ^{55}Fe source used to adjust the energy scale. Calibration curves with known amounts of ^{37}Ar and ^{39}Ar 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 ^{37}Ar 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): ^{81}Kr, 0.14 ± 0.10; ^{83}Kr, < 1.

Sample	Weight (g)	Tritium (dpm/kg)	Hydrogen (cm^3/g)	Helium (cm^3/g)	^{39}Ar (dpm/kg)	^{37}Ar* (dpm/kg)
			Apollo 11			
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		
			Meteorite(1)			
Sprucefield L4 (nonmagnetics)	(21)	290 ± 30		$<5 \times 10^{-3}$	7.7 ± 0.4	12.6 ± 1.4

*Extrapolated to 21 July 1969.