# The particle track record of the Ocean of Storms

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Abstract—In Apollo 12 rocks the numbers of tracks from the solar and galactic iron group cosmic rays imply surface residence times that range from < 10,000 years to  $\sim 30$  million years. The presence of steep track gradients at exposed surfaces shows that some rocks have been on the lunar surface in only one position, while others have been turned over and moved more than once. For example, rock 12017 was raised to within one meter of the surface, later thrown to the very surface, then flipped over and recently splattered with molten glass (just 9000 years ago). The abundance of nuclear interaction (spallation) tracks induced by the penetrating galactic protons provides residence times for different rocks in the top meter of soil of  $\sim 20$  to 750 millions of years. The erosion of lunar rocks is estimated by comparing the cosmic ray track distributions in lunar rocks with the one found in an uneroded glass detector exposed in Surveyor III. Erosion at a rate of about one atomic layer per year is inferred. By inducing uranium-235 fission tracks we have measured widely ranging uranium concentrations: less than  $10^{-3}$  parts per million in pyroxenes,  $\sim 1$  ppm in glass, and up to 170 ppm in zircon. The fossil track abundance in the zircon gives no evidence for the presence of extinct radioactivity by plutonium-244 or by super-heavy nuclei.

### Introduction

THE ABUNDANT particle tracks found in most lunar samples constitute a highly detailed record of the diverse chronology of lunar samples. Solidication ages are recorded by uranium-238 fission tracks (PRICE and WALKER, 1963a; FLEISCHER and PRICE, 1964a, b); times of exposure on the lunar surface (surface residence times) are given by tracks or iron group nuclei in the cosmic rays (CROZAZ et al., 1970a; FLEISCHER et al., 1970a, later PRICE and O'SULLIVAN, 1970; LAL et al., 1970) and we shall see that nuclear interaction (spallation) tracks (FLEISCHER et al., 1970a, b) measure the total time spent near and at the lunar surface.

## **PROCEDURES**

The primary new technique employed in this work is the use of spallation tracks to measure the total time near the lunar surface. We have found that spallation tracks yield ages that agree with radiometrically measured cosmic ray exposure times. For other procedures, including etchants used, our previous work (Fleischer et al., 1970b) should be consulted.

Technique for measuring spallation ages

Previously (FLEISCHER et al., 1970b) we demonstrated that the short, nearly featureless tracks (FLEISCHER et al., 1967) produced by nuclear interactions of penetrating primary cosmic ray particles increase in number with the time of exposure in the top 1–2 meters of soil. At the same time we noted that the observed density of these tracks varied appreciably from grain to grain, even though there is little variation with position in rocks of the sizes that have been available from Apollo 11 and Apollo

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12. The observed grain-to-grain variation in rocks is most likely a variation of etching efficiency that depends primarily (as judged by etching different portions of a fragmented crystal) on the crystallographic orientation of the etched surface and very likely on compositional variations, but only secondarily on the etching time, and very little (Fleischer et al., 1967) on the orientation relative to the incident cosmic ray nuclei. A reproducible measurement is made by etching a number of grains of the minerals of interest (in this case, pyroxenes) and choosing the highest track densities present as representative of the highest etching efficiency.

As long as this procedure is used in obtaining both the natural track counts ( $\rho_{\rm sp}$ ) and the calibration counts (P) that measure production rates, the ratio  $\rho_{\rm sp}/P$  will be reproducible on a given rock. Ideally the irradiation and the subsequent calibration would be done for the same crystals as the natural track counts.

The production rate P is measured for individual surface samples by bombarding annealed (track free after 17 hours at  $820^{\circ}$ C in platinum boats) samples with 3 GeV protons from the Princeton-Pennsylvania Accelerator in order to simulate the nuclear-active cosmic rays incident upon the moon. In reality these active particles include not only protons but primary helium and heavier nuclei and secondary high energy neutrons and pions. At the very surface the protons and alphas dominate. Although minor errors are introduced in calibrating with a single type of particle at a single energy, they are small relative to the variations inherent in the uncertainties of individual rock histories as to depths of burial and detailed geometries and compositions of shielding material. As judged by the etching of annealed samples following irradiation with fission fragments, the thermal treatment has not altered the registration properties of the pyroxenes.

### **RESULTS**

# Spallation ages and proton doses

In the preceding section we indicated how reproducible values are measured for the ratio of the spallation track density  $\rho_{\rm sp}$  to the production rate P of recoil tracks caused by high energy protons. The ratio  $\rho_{\rm sp}/P$  is the dose of protons needed to produce the observed spallation track density. In Table 1 measured values of  $\rho_{\rm sp}/P$  are given for a group of lunar samples in the column headed Proton Exposure. By assuming a flux  $\phi$  of  $3 \times 10^7/{\rm cm}^2$ -yr of primary cosmic ray nucleons (BAZILEVSKAYA et al., 1968) we calculate spallation Surface Ages given in the next column of Table 1 by  $\rho_{\rm sp}/P\phi$ , the spallation ages if the entire proton exposure occurred on the lunar surface.

For samples such as those listed, which were all picked up at the surface, these ages should be to first approximation directly comparable with the radiometrically measured spallation ages—listed in the right-hand column of Table 1. In general the agreement is excellent, well within the scatter among different radiometric ages where more than one such age is available. In a higher approximation the agreement between the radiometric age and the track spallation surface age depends on the detailed burial history of the rock. Different track and radiometric ages for samples with complex burial histories will result if the cross-sections for the individual relevant spallation reactions vary differently with depth of burial (Honda and Arnold, 1964). In addition, differences can arise if a sample is exposed at depth and then moved to the surface at a time that is recent with respect to the half life of the nuclide being used to measure the production rate radiometrically.

The true proton exposure ages are uncertain, however, because we do not know the samples' depths of burial during proton exposure. As accelerator experiments have shown (Honda and Arnold, 1964), a cascade of nuclear-active, secondary

Sample Number	Production rate (P)* (tracks/109 protons)	Observed track density $(\rho_{\rm sp})$ $({\rm cm}^{-2})$	Proton exposure $(\rho_{\rm sp}/P)$ (protons/cm <sup>2</sup> )	Surface age (10 <sup>6</sup> yr)	Minimum spallation age (10 <sup>6</sup> yr)	Radiometric spallation ages (10 <sup>6</sup> yr)
10017	1.7†	$2.1(\pm 0.1) \times 10^7$	$1.2 \times 10^{16}$	420	170	200-640(a,e,f,g,i,j)
10044	1.0	$8.2(+1.2) \times 10^{6}$		270	110	$56-100^{(f)}$
10049	2.39	$1.49(+0.15) \times 10^{6}$		21	8.5	$22.5-25^{(g,1)}$
12002	1.7†	$2.66(+0.25) \times 10^{6}$		55	20	50-145(b,d,h)
12017	1.45	$4.57(\pm 0.32) \times 10^{6}$		105	40	
12021	2.31	$5.1(+0.3) \times 10^{7}$		740	300	300 <sup>(h)</sup>
12065	1.35	$6.81(\pm 0.28) \times 10^6$		170	70	160-200(e,k)

Table 1. Track spallation ages of lunar pyroxenes.

protons and neutrons builds up with depth and then attenuates. From the data of FLEISCHER et al. (1967) we estimate that in order to produce optically visible, etched tracks from spallation recoil nuclei, reactions are required in which at least 5 nucleons are ejected from the struck nucleus. In such reactions the maximum flux is  $\sim 2.5$  times the primary proton flux and occurs at a depth of about 20 to 25 cm of soil (or 10 to 12 cm of rock) (Kohman and Bender, 1967). The column labeled Minimum Spallation Age in Table 1 gives minimum times corresponding to burial at a 25 cm soil-equivalent depth. It should also be evident that the observed track densities could have been produced by much longer exposures than we have listed, if the samples were located at greater depths where the high-energy particle flux is corresponding lower.

At present, track spallation ages are of relatively low precision. They do, however, give reasonable agreement with the radiometric ages, and they give track workers a new tool for assessing radiation exposure histories. So far, we report spallation results only for pyroxenes. However, by measuring spallation ages for another mineral with an identical track registration threshold, an improved internal check on the accuracy of such ages will be possible. By measuring ages using minerals of different thresholds and therefore different depth variations of the track production rate, data on depths of burial will be obtained. As we shall see, comparison of spallation track ages with surface residence ages calculated from the track densities from heavy cosmic rays also yields such data.

# Tracks of heavy cosmic rays: rock 12017

We have shown previously (CROZAZ et al., 1970; FLEISCHER et al., 1970a) how the dominant cosmic ray tracks from the iron-group nuclei can be used to measure the surface residence times of rocks and rock fragments, and how from steep track density gradients near space-exposed surfaces, former orientations of rocks can be inferred. As an example, the results shown in Fig. 1 for rock 12017 allow us to derive solely from track data the complicated and varied history given in Table 2.

<sup>\*</sup> Absolute values uncertain to  $\pm 30\%$ , but relative values are valid for 10049, 12002, 12017, 12021, and 12065.

<sup>†</sup> Average of other values.

<sup>&</sup>lt;sup>a</sup> Albee et al. (1970); <sup>b</sup> Alexander et al. (1971); <sup>c</sup> Bloch et al. (1971); <sup>d</sup> D'Amico et al. (1971); <sup>e</sup> Eberhardt et al. (1970); <sup>f</sup> Fireman et al. (1970); <sup>g</sup> Hintenberger et al. (1970); <sup>h</sup> Martiand Lugmair (1971); <sup>i</sup> Marti et al. (1970); <sup>j</sup> O'Kelley et al. (1970); <sup>k</sup> Stoenner et al. (1971); <sup>l</sup> From measurements by Funkhouser et al. (1970).

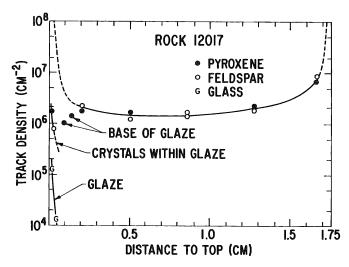


Fig. 1. Cosmic ray track distribution in rock 12017. The top of this rock was coated with glass of maximum thickness 0.15 cm. Tracks in crystals within the glass show it to be recently formed. Pyroxene and glass track densities are corrected for the measured etching efficiencies of 0.7 and 0.08.

Spallation tracks indicate the period over which the rock was exposed to galactic cosmic rays, and they are responsible for the first two entries in Table 2. The increases in cosmic-ray track density near both surfaces show that both sides have been exposed to space, and the slight asymmetry in the profile shows that the bottom received the longer exposure, roughly 1 million years, as compared to  $7 \times 10^5$  years for the top. At the very top is a glass coating that apparently was splashed on after the rock was positioned with that side up. From (presumably annealed) crystals trapped within the coating its space exposure is inferred to be only  $\sim 9000$  years, using the track production rate given by FLEISCHER et al. (1970b). The glass itself (microprobe analysis by wt. %: SiO<sub>2</sub>, 46.4; FeO, 17.7; Al<sub>2</sub>O<sub>3</sub>, 10.5; MgO, 11.0; CaO, 9.30; TiO<sub>2</sub>, 2.80) has the track retention characteristics given in Fig. 2. This glass is not highly retentive, allowing fading in 2 years at 400°K and 500 years at 350°K. We estimate that with the thermal cycling that occurs on the moon, tracks in a glazed rock with estimated peak temperature 360°K, would only be preserved over (very roughly) the last 500 years, as explained further in the caption to Fig. 2. We note parenthetically that in the glass there is a pronounced additional track fading that produces a decrease in track density toward the surface in the top 30  $\mu$ , a distance that corresponds to two or three optical depths for visible light. The cause of this effect has not been identified.

Table 2. Simplest track chronology for rock 12017.

Time (years before present)	Event		
up to ~ 105,000,000	Buried > 200 cm		
$\sim$ 105,000,000	Moved to $< 200 \mathrm{cm}$ and $> 15 \mathrm{cm}$		
$\sim 1,700,000$	Moved to surface		
$\sim 700,000$	Flipped over		
$\sim 9000$	Splattered with hot glass		
$\sim$ 500 to 0	Glass records solar flare particles		

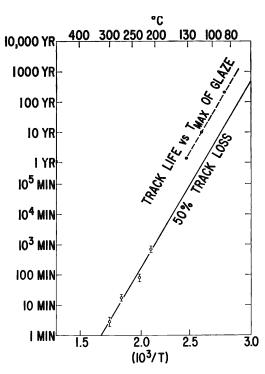


Fig. 2. Track retentivity in the glaze on rock 12017. Extrapolation of the data from lower temperatures predicts 50% track fading after one month continuously at 400°K. The dotted line indicates probable track life on the surface of the moon as a function of the maximum temperature reached by the glaze at lunar noon. Temperature vs. time data from the infra red measurements of SINTON (1962) were used.

In summary the low-energy cosmic rays (dominantly solar flare particles) have been recorded over different time intervals: the glaze over the last 40 to 50 solar cycles, the crystals within the glaze over the last  $\sim 800$ , and the bottom of the rock over a more ancient group of  $\sim 500,000$  cycles. Track distributions in these three sites should allow the proposed (PRICE and O'SULLIVAN, 1970) "solar flare paleontology," comparing ancient solar spectra at different periods of time.

## Surface residence times

Table 3 Summarizes cosmic-ray track information for Apollo 12 rocks and gives the most current data on 10049. This is an Apollo 11 rock of special interest because its surface time of 29 m.y. agrees with the 24 m.y. inferred from radioactivity measurements of spallation-produced nuclides (Funkhouser et al., 1970; Hintenberger et al., 1970) and the 21 m.y. inferred here from spallation tracks. In short this sample spent all of its near surface time directly exposed to space and underwent very little erosion (which would have lowered the track density). The limit on erosion ( $< 3 \times 10^{-8}$  cm/yr) is consistent with what we will infer shortly in this paper from our Surveyor III results. Table 3 shows a wide range of surface exposure times—from  $\sim 10^4$  to  $3 \times 10^7$  years—for samples some of which have been in a single surface position, some in at least two positions, and one in at least three.

The Surveyor III data has also made possible more reliable measurements of short surface residence times of small grains. Here the most abundant tracks are solar

Table 3. Minimum cosmic ray track densities and surface residence times for lunar samples.

Sample	Mineral	Track density (cm <sup>-2</sup> )	Depth in sample (cm)	Surface residence time (million years) (top/bottom)
10049	Pyroxene	$1.55 \times 10^{7}$	0.90 cm	(29 total)
12002	Pyroxene	$2.8 imes10^6$	5.0	(24/0)
12017	Feldspar	$1.51 \times 10^{6}$	0.45	(0.7/1.0)
12017	Feldspar in glaze	$8  imes 10^5$	0.02	(0.009/0)
12021	Pyroxene	$5.0  imes 10^6$	4.0	(13/13)*
12065 12025)	Pyroxene Pyroxene)	$2.2 \times 10^6$	6.4	(14/0)
12028 (Soil)	Feldspar	$3 \times 10^7 \dagger$		(110 total)‡
12025, 4, 54–8.5, 9, 9	Pyroxene	$5 \times 10^7$	0.002	0.01/0)§

<sup>\*</sup> This result disagrees with that of PRICE (personal communication); a mix-up in sample position designation (either his sample or ours) is suspected.

† Average of  $100 \mu$ – $400 \mu$  diameter grains.

heavy cosmic rays whose flux previously was highly uncertain. Because of the presence in most lunar samples of an unknown amount of erosion, the track production rate vs depth was only a lower limit. However, with the recent data (adjusted to solar cycle average) for the uneroded Surveyor III glass and the assumption that it represents a typical solar cycle, the ages can be computed for small grains such as that sketched in Fig. 3. The steep track gradients mapped around roughly half of its perimeter reveal that at one time this grain was exposed directly to space, as a small grain resting on what is drawn as its right side. By matching the steepest gradient in this grain with calculations based on the Surveyor III flux of solar heavies (Fleischer et al., 1971) a surface residence time of 10,000 years is obtained. This age would be altered somewhat if the solar fluxes are used that have been inferred by Crozaz and Walker (1971) or Price et al. (1971) from the same material. Hence, the absolute surface time for this sample is subject to possible revision, but is roughly correct and clearly much shorter than the 0.5 to 30 m.y. that typifies most larger moon rocks.

## Burial and burial depths

By comparing track spallation ages with track surface residence times and with radiometric spallation ages, permissible burial depths can be inferred. Thus for rock 10049, where all three agree, the entire exposure must have been at the surface. For the other rocks listed, where the surface residence age is shorter than the spallation age, the samples must have been buried over most of their spallation exposure times.

## Lunar erosion

In a separate experiment (Fleischer et al., 1971) using tracks etched in a glass optical filter from the Surveyor III television camera we have measured the energy spectrum of the iron group solar cosmic ray particles over the energy range 1 to 100 Mev/nucleon, finding for the differential flux  $1.8 \times 10^3 \text{ E}^{-3}$  particles/m<sup>2</sup>-secstr-MeV/nucleon. We have also observed high energy fission of Pb, induced by galactic cosmic ray protons and alpha particles.

<sup>‡</sup> Average time in top 60 cm of soil, calculated in same manner as in Fleischer et al. (1970b).

<sup>§</sup> Using solar spectrum from Surveyor III glass (Fleischer et al., 1971) after adjustment for solar cycle.

SAMPLE 12025,4,54 - 8.5,9,9

CONTOURS IN 10<sup>8</sup> TRACKS/CM<sup>2</sup>

TRANSMISSION MICROGRAPH COUNT
OPTICAL COUNT x 3

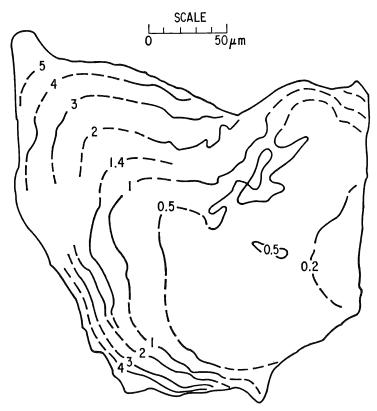


Fig. 3. Cosmic ray track density distribution in a grain found at 8.5 cm depth in core 12025. The gradients identify the surfaces exposed directly to space and to heavy cosmic ray nuclei from the sun. The surface exposure was for  $\sim 10,000$  years.

Using this energy spectrum and the track density profiles measured in lunar rocks we have obtained an estimate of the rate of fine scale erosion on the moon. Making the necessary corrections for the different properties of the glass and the lunar rocks, for the different solid angles involved, for the solar cycle variation, and assuming the present solar cycle to be typical of the last few million years, we find an erosion rate of 0 to  $2 \times 10^{-8}$  cm/yr to be consistent with the track profiles measured by four groups on rocks 10017 and 10003. The much lower track density profile of rock 10058 is consistent with the recent removal of a chip of appreciable thickness and does not contradict our remarkably low fine-scale erosion rate. Similar results have simultaneously been obtained by Crozaz and Walker (1971) and Price et al. (1971).

## Uranium contents and fission track dating

Since fission track dating requires the presence of uranium (PRICE and WALKER, 1963b), the induced fission track measurements given in Table 4 are relevant. In the cases shown, uranium is too low to allow fission track dating of any of the samples except for zircon LZ where an upper limit can be given. Since the fossil track content

Mineral	Sample	Uranium* (wt fraction × 10°)	Notes
Augite	12017,17,6,3	0.4 (±60%)	Not including visible inclusions
Augite	12017,17,6,3	1.5 (±30%)	Including visible inclusions
Augite	12021,1,4,5	1.0 (±50%)	Not including visible inclusions
Augite	12021,1,4,5	4.5 (±20%) <b>©</b>	Including visible inclusions
Augite	12065,6,6	0.5 (±70%)	Including visible inclusions
Glass	12017,8,6	$1,230 (\pm 8\%)$	
Zircon† Zircon†	LZ (from Apollo 11 fines Z-2 (from Apollo 11 fines)	$167,000  (\pm 12\%) \ < 10,000$	90% confidence

Table 4. Uranium content of lunar samples.

was 1 to  $3 \times 10^8/\text{cm}^2$ , the ages would be 1.3 to  $3.3 \times 10^9$  years if these were all fission tracks, and less if an appreciable fraction were of other origin. There is thus no evidence for an excess of fission tracks from presently extinct fission activity by Pu-244 or super-heavy elements.

### Conclusions

Track spallation measurements and solar cosmic ray tracks in the Surveyor III glass add two new dimensions to lunar information available from track measurements. By comparing the spallation and surface residence ages inferred for individual rocks (using spallation and cosmic ray tracks), surface and near surface chronologies can be constructed. By comparing track gradients and abundances in the uneroded glass and in eroded rocks, erosion rates for individual rock surfaces can be determined. Not surprisingly, individual rock histories vary widely.

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<sup>\*</sup> Tracks observed on interior surfaces, except as noted.

<sup>†</sup> Tracks observed in Lexan adjacent to sample.

 $<sup>\</sup>bullet$  Most uranium-rich inclusion scanned contained  $\sim 3 \times 10^6$  atoms of uranium.

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