Reports

Extralunar Dust in Apollo Cores?

Abstract. Densities of nuclear tracks exceed 10^{11} per square centimeter in several percent of the micrometer-size silicate grains from all depths in the 12and 60-centimeter lunar cores. Either these grains were irradiated in space as extralunar dust or the ratio of iron to hydrogen in low-energy (about 1 million electron volts per nucleon) solar particles is orders of magnitude higher than in the photosphere.

With the Berkeley 650-kv electron microscope we have found track densities up to and exceeding 3×10^{11} per square centimeter in small silicate crystals (diameter ~0.5 to ~3 μ m) from all depths in Apollo 11 and 12 cores. We will discuss the constraints that these remarkable observations place on the origin of the lunar dust.

By diffraction contrast, tracks are visible in silicate grains (Fig. 1) but must be observed through an amorphous layer (~ 1000 Å thick) that is sug-

gestive of solar wind radiation damage. We find that a very light etch removes this layer and improves track visibility. A carbon film is evaporated onto a microscope slide containing grains of less than $\sim 3 \ \mu m$, is stripped off the slide, and is then floated on a solution of 2HF : $1H_2SO_4$: $280H_2O$ for 30 seconds. In addition to serving as a support for the tiny grains, the film replicates their original shapes, so that the decrease in grain diameters resulting from etching can be followed. This very



Fig. 1. Unetched crystal of clinopyroxene containing 7×10^{30} track/cm² which are attributed to nuclei with atomic number $Z \approx 26$ emitted in solar flares. This crystal, like those in Fig. 2, was selected from the residue remaining on a glass slide after part of an Apollo 12 core sample was poured onto the slide and then poured off (\times 73,500).

weak etch solution satisfactorily reveals tracks in all the common lunar silicates —pyroxenes, feldspars, and olivines without destroying glassy grains or other minerals.

In Fig. 2 etched tracks in pyroxene and olivine as identified by electron diffraction are shown. The slitlike shapes of the etched tracks in Fig. 2B are characteristic of olivine (1). The shapes of the etched lines, their angular and length distributions, the occasional gradients across a grain diameter, their annealing behavior (no tracks being visible either with or without etching after heating to $\sim 500^{\circ}$ C), and the absence of such lines in nontrack-recording crystals (ilmenite, for example) identify them as nuclear tracks (2). Other kinds of defects in lunar grains will not be discussed here.

From our observations of well over 1000 etched grains, we estimate that, at all depths, at least 15 percent of silicate grains with diameters of 0.5 to 3 μ m have track densities exceeding 1010 per square centimeter. About twothirds of the grains are glass (which must be etched under different conditions) and nontrack-recording minerals, and they are excluded from this analysis. At least 3 percent of the 0.5- to $3-\mu m$ silicate grains at all depths have $>10^{11}$ track/cm². The percentage is probably even higher, but track densities greater than $\sim 4 \times 10^{11}$ per square centimeter are simply not resolvable.

X-ray powder patterns of unetched 0.5- to $3-\mu m$ grains, taken by H. R. Wenk, provide evidence of severe strains and support our contention that the grains have been severely damaged by radiation (3).

Previous workers, who studied much larger grains (diameter $\geq 100 \ \mu m$) with optical microscopy (4-6), scanning electron microscopy (4), and a replication technique (5), found a wide distribution of track densities ranging from $< 10^6$ per square centimeter up to their limit of resolution, $\sim 3 \times 10^9$ per square centimeter, at all depths in the 10.5- and 12.5-cm Apollo 11 cores (4-6). With optical microscopy, we find that the fraction of large ($\gtrsim 50$ μ m) silicate grains containing > 10⁸ track/cm² slowly decreases from ~ 95 percent at the surface to ~ 75 percent at a depth of 60 cm.

We believe, with previous workers (4-7), that many of the grains with track densities up to a few times 10^9 per square centimeter could have been chipped off surface rocks by meteorite

impact and could have accumulated their tracks while residing on the surface before being stirred into the soil by later impacts.

Our main purpose in this report, however, is to point out the difficulty of accounting for small grains with track densities of $\ge 10^{10}$ per square centimeter throughout a depth of 60 cm. The maximum track density expected from either spontaneous or induced fission of uranium or of an extinct element, or from galactic cosmic rays or their spallation recoils, falls orders of magnitude short of that observed. The most reasonable origin of the tracks is irradiation by heavy nuclei emitted by the sun with energies of ~ 1 Mev per nucleon. This is about 10^3 times the average energy of solar wind ions and is probably associated with solar flares. Studies of solar flare tracks (4, 7, 8) and of flare-induced radioactivity (9) in lunar rocks with measured surface residence times of $\sim 10^7$ years strongly suggest that the average level of solar activity has not changed drastically over the last 10⁷ years. By using either an average energy spectrum $\propto E^{-3}$ [suggested by recent satellite measurements (10)] or an average rigidity spectrum $\propto e^{-R/100}$ Mv, we reach the same conclusion that a grain within ~ 10 μ m of the surface will accumulate 10¹¹ track/cm² in about 10⁷ years. At a depth of 100 μ m the required irradiation time is greater than 10⁸ years. Because of the steepness of the solar particle energy spectrum, the most rapid accumulation occurs by exposing each grain in the top 10 μ m of soil at least once. To irradiate 10 percent of the grains in the top 60 cm of soil with 10¹¹ track/cm² would require more than 6×10^{10} years at the present level of solar activity, if the Fe/H ratio in solar particles and in the photosphere is assumed to be the same, $\sim 4 \times 10^{-5}$. Since there is no indication of a decrease of track density with depth, a much greater thickness than 60 cm presumably contains high track densities; and an even longer irradiation time would be required to produce them.

In order for the grains to have acquired their high track densities while on or near the lunar surface in a time less than the age of the solar system, either we must postulate that the irradiation occurred at an early stage in the evolution of the sun, when its power output in solar flares was orders of magnitude higher than it has been

29 JANUARY 1971

for the last $\sim 10^7$ years, or we must postulate that the Fe/H ratio in lowenergy solar particles is several orders of magnitude higher than in the photosphere.

The difficulty disappears if the tracks accumulated in grains that orbited the sun but gradually spiraled in, owing to the Poynting-Robertson effect, until they were swept up by the moon and built up a lunar soil. Although grains that collide with rocks would be destroyed (giving rise to glass-lined craters), those that impinge with minimum velocity (~2 km/sec) on a fluffy, porous soil of like grains can transfer much of their energy to those grains, and portions of them should survive. Stored tracks are known to be unaffected by shock pressures up to at least 150 kb, which corresponds to impact velocities considerably higher than 2 km/sec.

Grains of initial radius 100 μ m and initial perihelion 2.8 A.U. (corresponding to the asteroidal belt) would have a Poynting-Robertson lifetime of 2×10^6 years if they were perfectly absorbing or reflecting and would have a considerably longer lifetime if they were transparent. In 10⁶ to 10⁷ years they would accumulate 10¹⁰ to 10¹¹ track/cm² in their outer 20 μ m or so. Since the Poynting-Robertson lifetime is proportional to radius, very small grains could not accumulate high track densities, and we would attribute the highest track densities to fragments from the outer portions of grains of radius $\gtrsim 100 \ \mu m$ that fractured on impact.

Our observations do not allow us to distinguish whether high track density grains have been added to the soil on a continuing basis or only during the early history of the moon. If addition has continued, we would expect the mineralogy and chemistry of those grains and of meteorites to be similar. From a gross analysis of enriched concentrations of certain trace elements, Keays et al. (11) conclude that ~ 1 percent of the lunar soil is of meteoritic origin. It would be interesting to see whether the meteoritic fraction is larger for small grains of diameter $< 3 \mu m$. It would also be interesting to search Pacific Ocean sediments for grains with high track densities, because their existence would provide evidence of a continuing infall.

The concentration of rare gases of solar origin in the lunar soil is an inverse function of grain size (12-14). The flux of solar wind gases, which penetrate less than ~ 1000 Å below the surface, is so high that the gas concentration near the surfaces of grains quickly reaches a saturation level in a time less than $\sim 10^3$ years (13) and



Fig. 2. Etched crystals. (A) Clinopyroxene containing 3×10^{11} track/cm² from Apollo 12 core; etched for 30 seconds in 2HF : 1H₂SO₄ : 280H₂O (× 136,500). (B) Olivine containing 1.2×10^{9} track/cm² from Apollo 12 core; etched for 1 minute in 2HF : 1H₂-SO₄ : 45H₂O (× 23,800).

thus does not provide a test of the hypothesis of extralunar origin. A selective study of the more deeply seated gases produced in solar flares might, however, provide such a test. By leaching away the surfaces of specific silicate grains, one might be able to study the solar flare gas content of the interior portions. However, because of possible diffusion effects, a rare gas study would be more difficult to interpret than our track evidence.

Spallogenic nuclides are produced by cosmic-ray bombardment down to a depth of ~ 1 m. From analysis of stable spallogenic rare gases, various groups have inferred an exposure age of $\sim 5 \times 10^8$ years for the soil (13-15). This is not its absolute age but refers to the time spent within $\sim 1 \text{ m}$ of the surface. Provided deep stirring has occurred, this age can be compatible either with an early origin of the soil or with continuing production.

An extralunar origin of the grains with high track densities is consistent with Gold's dust model (16) of the origin of the lunar surface. We believe that the attractive features of his model outweigh the difficulties. An important and testable consequence of the model is that the grains with high track densities should also occur at much greater depths than 60 cm, perhaps orders of magnitude deeper. The desirability of a 4-m core sample such as had been planned on Apollo 13 is obvious.

The most popular model of the origin of the soil is that a regolith accumulates on a solid bedrock by comminution of chips of the bedrock resulting from meteorite impacts (17). Our explanation for the high track densities could be compatible with this impact model only if the extralunar infall had continued over several billion years, so as to maintain a large fraction of infallen grains with very high track densities. We believe that the simplest explanation of our results is that much of the soil developed from infall of extralunar particles (18).

Note added in proof: We have now directly measured the energy spectrum of Fe nuclei over a 2.5-year period by analyzing tracks recorded in a glass filter in the Surveyor 3 camera that was brought back from the moon by the Apollo 12 astronauts. We have found that the Fe/H ratio in lowenergy solar particles is much higher than we had assumed from the photospheric ratio. The high Fe flux reduces but does not entirely eliminate the

References and Notes

difficulty of accounting for the grains

with high track density. An extralunar

origin of part of the soil is still con-

D. J. BARBER*

I. HUTCHEON

P. B. PRICE

sistent with our observations.

- 1. M. Maurette, P. Pellas, R. M. Walker, Na-M. Madette, T. Tehas, K. M. Walker, Nature 204, 821 (1964).
 Etched nuclear tracks of natural origin were
- first seen by P. B. Price and R. M. Walker [Nature 196, 732 (1962)], who observed spontaneous fission tracks in terrestrial micas by electron microscopy. Etched tracks of heavy cosmic rays in extraterrestrial bodies (meteor ites) were first observed by optical microscopy and reported by Maurette et al. [see (1)] of the x-ray analysis will be 3. Further details
- reported elsewhere.
- reported elsewhere.
 G. Crozaz, U. Haack, M. Hair, M. Maurette,
 R. Walker, D. Woolum, in *Proceedings of the Apollo 11 Lunar Science Conference* (Pergamon, New York, 1970), vol. 3, p. 2051.
 R. L. Fleischer, E. L. Haines, H. R. Hart,
 R. T. Woods, G. M. Comstock, *ibid.*, p. 2103.
 D. Jol D. Modauroull, J. Willinging, G.
- D. Lal, D. Macdougall, L. Wilkening, G. Arrhenius, *ibid.*, p. 2295.
 P. B. Price and D. O'Sullivan, *ibid.*, p. 2351.
 D. J. Barber and P. B. Price, in preparation.

- 9. J. R. Arnold, private communication. 10. J. Hsieh and J. A. Simpson, private communication.
- R. R. Keays, R. Ganapathy, J. C. Laul, E. Anders, G. F. Herzog, P. M. Jeffery, *Science* 167, 490 (1970).

- 12. D. Heymann, A. Yaniv, J. A. S. Adams, G. E. Fryer, *ibid.*, p. 555. 13. P. Eberhart *et al.*, *ibid.*, p. 558.
- 14. T. Kirsten, F. Steinbrunn, J. Zähringer, ibid.,
- I. Kirsten, F. Steinbrunn, J. Zanringer, *ibid.*, p. 571.
 K. Marti, G. W. Lugmair, H. C. Urey, *ibid.*, p. 548; R. O. Pepin, L. E. Nyquist, D. Phin-ney, D. C. Black, *ibid.*, p. 550; C. M. Hohen-berg, P. K. Davis, W. A. Kaiser, R. S. Lewis, J. H. Reynolds, in *Proceedings of the April of L1 Jures Conference Conference*. Apollo 11 Lunar Science Conference (Perga-
- mon, New York, 1970), vol. 2, p. 1283.
 16. T. Gold, "The Nature of the Surface of the Moon" (preprint 380, Cornell University Center for Radiophysics and Space Research, Ithaca, N.Y., 1970); _____, Proc. Amer. Phil. Soc., in press; -169, 1071 (1970). - and S. Soter, Science
- 17. E. M. Shoemaker et al., in Proceedings of the Apollo 11 Lunar Science Conference (Pergamon, New York, 1970), vol. 3, p. 2399. After the manuscript of this report was com-
- pleted, we learned of high voltage electron pleted, we learned of high voltage electron microscope observations of lunar crystals with track densities up to 10th per square centi-meter [J. Borg, J. C. Dran, L. Durrieu, C. Jouret, and M. Maurette, *Earth Planet. Sci. Lett.* 8, 379 (1970)]. They used an acid solu-tion of the sume comprovimed by 70 times as tion that was approximately 70 times as strong as the one we used, but they reported that the tracks in their grains were not etched. is possible that all silicates were completely dissolved in their solution, leaving only min-eral grains that would require a different chemical etchant. One of their photographs was of a cristobalite grain, which we believe would not be etched in their solution.
- We are greatly indebted to F. Borden for ex-perimental assistance. Supported by NASA contract NAS 9-10488 and by AEC contract 19. AT(04-3)-34.
- On leave from Department of Physics, Essex University, Colchester, U.K.
- 8 October 1970

Ordering of V^{2+} , Mn^{2+} , and Fe^{3+} lons in Zoisite, $Ca_2Al_3Si_3O_{12}(OH)$

Abstract, The presence of very small amounts of Mn^{2+} , V^{2+} , and Fe^{3+} ions in zoisite can be easily detected by the electron paramagnetic resonance technique at room temperature. The Mn^{2+} and Fe^{3+} ions are completely ordered and are probably located in the Ca(1)- and Al(II)-sites, respectively, whereas the V^{2+} ions probably occupy both Ca(1)- and Ca(2)-sites, with a preference for the Ca(1)-site.

The electron paramagnetic resonance (EPR) technique, primarily used thus far in the field of solid-state physics, has been applied only recently to mineralogical problems. The EPR technique can be used to detect very small

amounts of transition metal impurities [in the parts-per-million (ppm) range] in their various valence states in a diamagnetic mineral host. From the variation of the EPR spectra as a function of the crystal orientation within the

Table 1. Spin Hamiltonian parameters in zoisite.

Ion	g	D (cm ⁻¹)	E (cm ⁻¹)	Average $ A $ (cm ⁻¹)	Site sym- metry	Mirror plane	Angle* between principal and crystallographic axes
Mn ²⁺	$\begin{array}{c} 2.003 \pm 0.005,\\ \text{isotropic} \end{array}$	$(103 \pm 5) \times 10^{-4}$	$(34 \pm 2) \times 10^{-4}$	$(85 \pm 2) \times 10^{-4}$, isotropic	т	(010)	5° ± 1°
V_1^{2+}	1.940 ± 0.007 , isotropic	< 0.014	< 0.005	$(88 \pm 4) \times 10^{-4}$, anisotropic	m	(010)	$7^{\circ} \pm 2^{\circ}$
V_2^{2+}	$1.940 \pm 0.007,$ isotropic	< 0.014	< 0.005	$(100 \pm 6) \times 10^{-4}$, anisotropic	1	None	
Fe ^{s+}	$g_z = 1.96 \pm 0.01$	0.14 ± 0.03	<< D	None	т	(010)	$44^{\circ} \pm 1^{\circ}$

* The two coordinate systems are related to each other by a rotation about the b-axis.