The Radiation History of Material Returned by the Soviet Automatic Stations Luna 16 and Luna 20, According to Track Studies

L. L. Kashkarov, L. I. Genayeva, and A. K. Lavrukhina,
V. I. Vernadskiy Institute of Geochemistry
and Analytical Chemistry,
Moscow, U.S.S.R.

Fission tracks formed by the vH (very heavy) nuclei group ($23 \overset{\sim}{<} Z \overset{\sim}{<} 28$) of solar and galactic cosmic rays have been studied in silicate minerals of the lunar regolith returned by the Luna 16 and Luna 20 unmanned spacecraft. It is shown that the material in the Luna 16 core sample, from a typical mare region of the lunar surface, has undergone stronger irradiation by cosmic rays than material returned from a highland region by Luna 20. A low-irradiation component (about 10 percent of the total number of crystals) has been found in the Luna 20 core sample materials, which can possibly be attributed to material added to the main bulk of the regolith in the formation of the crater Apollonius C. From the track density distribution of crystals, as a function of depth in the regolith core sample, it follows that the process of formation of the upper layer of the regolith, both for the lunar mare and for the highland region, includes sequential layering of finely crushed crystalline matter and subsequent mixing of it by micrometeorite bombardment. A portion of the crystals with a very high track density ($\overset{\sim}{>} 10^9$ cm^{-2}) may be a component added to the lunar surface from outer space.

Regolith core samples returned to Earth by Luna 16 and Luna 20 differ mainly in that the former is lunar soil from a mare region (Mare Fecunditatis) of the lunar surface (ref. 1) and the second is typical highland soil taken from the region between Mare Fecunditatis and Mare Crisium (ref. 2). Study and comparison of the history of formation of the material of these two regions on the lunar surface is of special importance for studying the processes that formed the lunar surface.

In the last decade, the use of microdestruction of crystal structures by the heavy nuclei of cosmic rays has become one of the most effective methods of studying the cosmic history of extraterrestrial material (ref. 3). In this case, areas of microdestruction in silicate minerals along the tracks of heavy nuclei with charge $Z \overset{\sim}{>} 23$, can be detected in the form of hollow channel-tracks. The tracks can be observed after appropriate chemical etching of the sample surface to be studied has been made by a scanning electron or optical microscope (refs. 4 and 5) as well as by a high-voltage electron microscope (ref. 6).

The present work reports the results of fission track studies in various minerals separated from the entire depth of the lunar regolith cores returned by Luna 16 and Luna 20 in order to compare the radiation histories of the upper layer of the lunar regolith taken from two different regions of the lunar surface. Observation and measurement of the basic characteristics of the tracks were accomplished by means of scanning electron and optical microscopes. The technique allowed a count to be made of track densities (ρ_{sem}, ρ_{opt}) at 10^4 and 10^3 magnification, respectively, as well as measurements of the length and angular distribution of the tracks of the vH group nuclei of solar and galactic cosmic rays.

Crystals of olivine, feldspar, and pyroxene, 100–200 μm in size, were mounted in tablets of epoxy resin which were used to make polished sections and microsections and to enable subsequent etching. Control of mineralogical identification of the individual samples and the track etching efficiency under various conditions was accomplished by etching artificially induced tracks of ^{252}Cf fission fragments. Among the Luna 16 core samples studied, 50 percent were olivine, 30 percent were feldspar, and 20 percent were pyroxene. The Luna 20 regolith core was represented by olivine (8 percent), feldspar (67 percent), and pyroxene (25 percent). Chemical etching of the olivine crystals was carried out for periods of 30 to 240 minutes by the method developed by Krishnaswami (ref. 7). Feldspar and pyroxene crystals were etched in a solution of 3g NaOH and 4g H_2O, for a period of 4 to 8 minutes and 20 to 60 minutes, respectively (ref. 8).

Up to the present time we have studied a total of 160 crystals from the Luna 16 regolith core and 294 crystals from the Luna 20 regolith core. It should be noted here that the track densities observed in different minerals may differ because of differences in their sensitivity for recording nuclei with $Z \gtrsim 20$, as indicated by Bhandari (ref. 5). It was found that the density of tracks formed in lunar rock at a depth of about 100μm was approximately the same for feldspar and pyroxene ($\rho_{\text{feld}} = \rho_{\text{px}}$), and that the

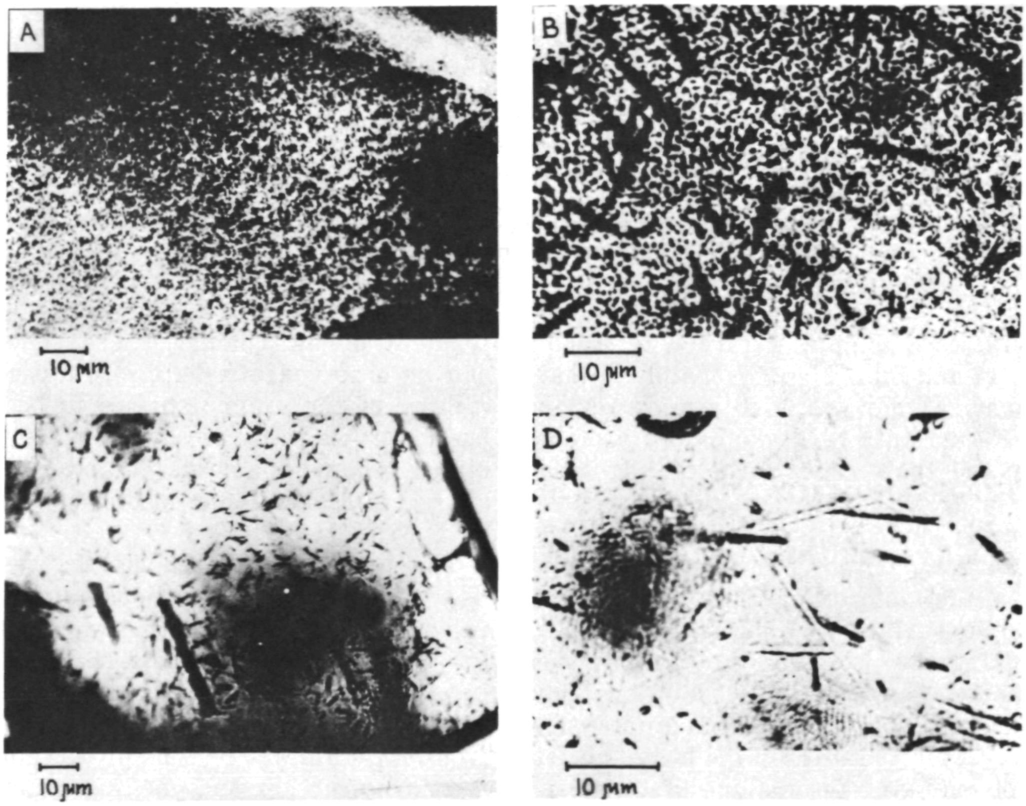


Figure 1.—*Microphotographs of tracks observed under various examination conditions on etched surfaces of feldspar crystals from the Luna 20 core: A—scanning electron microscope* $\rho \gtrsim 10^8$ *cm*$^{-2}$*; B—optical microscope, with long tracks seen on a background of short tracks* $\rho_{\text{opt}} \gtrsim 10^7$ *cm*$^{-2}$*; C, D—optical microscope, transmitted light, tracks with density* $\rho_{\text{opt}} \gtrsim 10^6$ *cm*$^{-2}$ *and* $\rho_{\text{opt}} \lesssim 10^6$ *cm*$^{-2}$*.*

track density ratio $\rho_{feld}/\rho_{ol} \simeq 2$, which characterizes olivine (ρ_{ol}) as the least sensitive track detector among the minerals studied. For our study, in a comparison of the results obtained, we allowed for the mineralogical composition of the regolith core samples—in particular, for the fact that 50 percent of the Luna 16 core samples studied were olivine, while olivine amounted to less than 10 percent in the Luna 20 core sample.

In the fission track studies, the principal attention was given to determining the extent of irradiation for the individual crystals, which was determined from the track densities observed in them. In this case, for estimation of the contribution of the vH group nuclei of solar and galactic cosmic rays to the track density, we used the following criteria:

1. The presence of a track density gradient in the interior of the crystals, related completely to the vH group of solar cosmic rays ($E_{kinetic} \lesssim 10$ MeV/nucleon), uniquely indicates irradiation of these crystals on the surface of the regolith, without any protective layer of material.
2. Excess values of the observed track densities $\rho_{sem} \gtrsim 10^8$ cm^{-2} also assign the sample of those irradiated by the vH group nuclei solar cosmic rays in the upper ($\sim$ mm) layer of the lunar regolith.
3. Crystals with track density $\rho_{sem} \lesssim 10^8$ cm^{-2} may contain a considerable portion of tracks, formed by the vH group nuclei of galactic cosmic rays, whose relative contribution increases in proportion to the decrease in observed track densities. On the basis of this dependence of track density on irradiation conditions, it becomes evident that crystals with track density of not over $\sim 10^6$ cm^{-2} contain traces of the effects of little but the heavy nuclei of galacic cosmic rays, and that they can be used to estimate the effective irradiation time in a layer of the regolith at a specific depth. In this case, the contribution of fragments from spontaneous fission of ^{238}U, whose concentrtion is not over 10^{-8} g/g, is only $\sim 10^4$ tracks per cm^2.

A comparison of the track densities observed by means of the scanning electron (ρ_{sem}) and optical (ρ_{opt}) microscopes was made on a large number of crystals in the ρ_{sem} range from $\sim 10^6$ to $\sim 10^9$ cm^{-2}. Examples of microphotographs of tracks observed under various conditions are presented in figure 1. Figure 2 is a microphotograph of a section of feldspar crystal (Luna 20), with a clearly expressed track density gradient and the corresponding histograms of change in track density with depth in the sample, as obtained by both optical and scanning electron microscopes (ref. 9). As seen from a comparison of the histograms, a similar distribution of track density with depth in the crystal is given by the data from the optical and scanning electron microscopes. However, under our conditions of etching and examination, ρ_{sem} — ρ_{opt} for $\rho_{sem} \lesssim 10^8$cm^{-2}. With increasing track density, the ratio ρ_{sem}/ρ_{opt} changes from two to five times, with increase in track density ρ_{sem} from 1.2×10^8 cm^{-2} to 5×10^8cm^{-2}. For large values of ρ_{sem} ($\gtrsim 5 \times 10^8$ cm^{-2}), the divergence between ρ_{sem} and ρ_{opt} increases.

Results

TRACK DENSITY GRADIENT IN INDIVIDUAL CRYSTALS

As known (ref. 10), observation of the track densities due to the vH group nuclei of cosmic rays, at various depths in crystals, is determined by the flux, free path length, and energy spectrum of the nuclei forming the tracks. Moreover, under irradiation conditions similar to those of the lunar surface, change in track density with depth in individual crystals also depends on their size, extent of erosion, and depth of occurrence in the regolith layer. In connection with this, the magnitude of the track density gradient, defined by the degree of change in track density with depth in the crystals, depends on many factors that cannot be quantitatively accounted for. However, in the study of crystals with maximum track density gradient values, it appears possible to obtain a quantitative characterization of the energy spectra

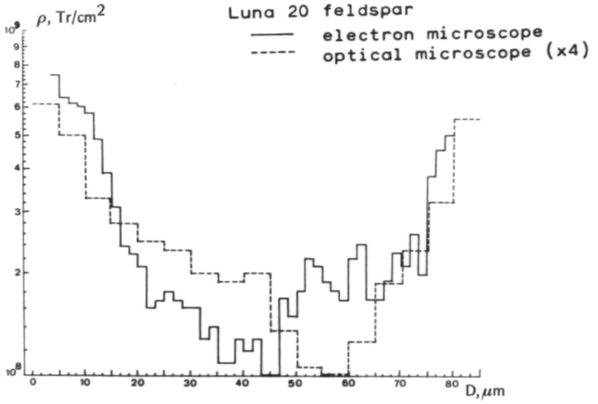
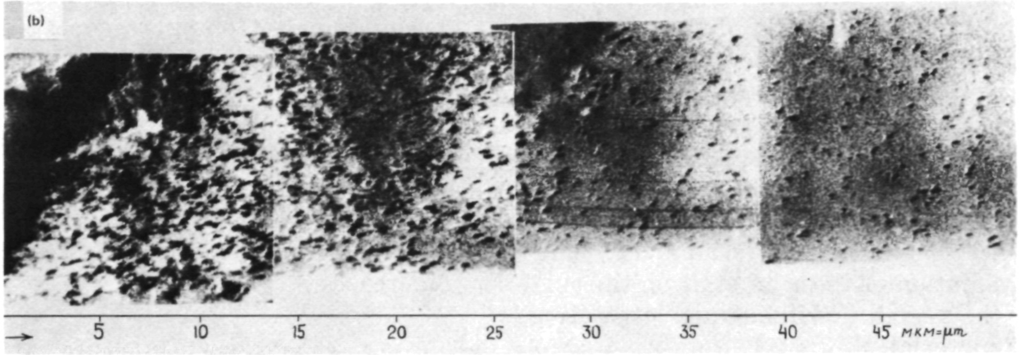


Figure 2.—*Track density gradient in feldspar crystal from the Luna 20 core from scanning electron and optical microscope data.*



of the heavy nuclei of solar cosmic rays. Moreover, as was pointed out above, we use the fact of the presence or absence of the gradient to estimate the degree of irradiation of the regolith material, a specific part of which was subjected to irradiation on the lunar surface without any protective layer.

Of the total number of crystals from the Luna 16 and Luna 20 regolith core samples studied up to the present time, the fraction of samples with a track density gradient is 30 percent and 5 percent, respectively. The magnitude of the gradient for all samples varies from a twofold to a tenfold change in track density at a depth of 100μm (see figs. 2 and 3). The exponent γ, corresponding to the maximum values of the track density gradient, in the formula for the differential energy spectrum of the vH group nuclei, $J = \text{const} \cdot E^{-\gamma}$, is 1.5 to 2 (ref. 11) which is in agreement with other results (refs. 12, 13, and 14).

The results of the measurement of track density gradient which we obtained showed the following:

1. The relative number of crystals with a track density gradient for the Luna 16 soil is six times that for the Luna 20 core sample, which can be explained by the difference in formation processes for the upper layer of the regolith in the lunar mare and highland regions. In the first case, formation of the upper layer of the regolith, owing to the level terrain, could have taken place by filling and mixing of thinner layers, while in the second case, owing to the presence of highlands, the growth of the regolith layer could have occurred more intensely. Furthermore, the exposure age of the regolith in the mare region of the Moon may be greater on the average than that in the highlands, where formation of the upper layer of the regolith could have occurred at a later time by accumulation of finely crushed material, as a result of meteorite bombardment of nearby bedrock. The

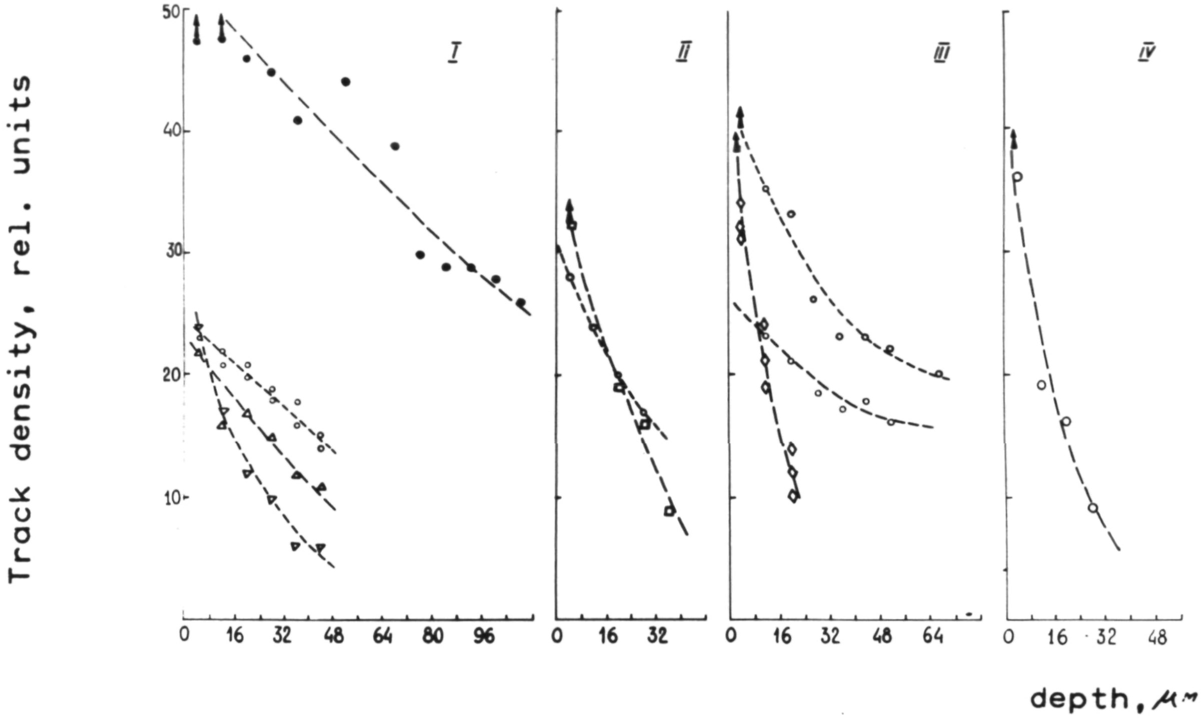


Figure 3.—*Examples of track density gradients observed in various crystals from the Luna 16 and Luna 20 cores.*

limits of change in track density gradients were the same for both regolith core samples.

2. The size of the gradient in individual crystals does not depend on the absolute track density or the mineralogical composition of the crystals.

Among the crystals having track density gradients, a small number (~ 1 percent) is encountered in which the track densities change from the surface inward from practically all sides. Examples of two such crystals of feldspar from the Luna 20 core sample are presented in figure 4. This track density distribution with depth in the crystals may be the result of uniform irradiation of the crystals from all sides, or of their repeated displacement on the surface of the lunar regolith, or of irradiation of these samples in space. The predominant fraction of the crystals has only a local, one-sided track density gradient, which also can be considered from two points of view: either these crystals were irradiated under 2π-geometry, because of being on the surface of the lunar regolith; or they are fragments of larger ($\gtrsim$ mm) primary objects, irradiated in the free state before falling to the lunar surface and being broken up after this irradiation.

DISTRIBUTION OF CRYSTALS ACCORDING TO THEIR TRACK DENSITY

The results of track density measurements (ρ_{opt}) in crystals separated from various depth zones of the Luna 16 and Luna 20 regolith cores are presented in table 1 and as histograms in figure 5. On the basis of distribution of the crystals studied according to their track densities, three groups can be distinguished with the following ranges of track density: I, $\rho_{opt} \lesssim 10^6$ cm^{-2}; II, $10^6 \lesssim \rho_{opt} \lesssim 10^7$ cm^{-2}; and III, $\rho_{opt} \gtrsim 10^7$ cm^{-2}. Comparison of the data obtained for Luna 16 and

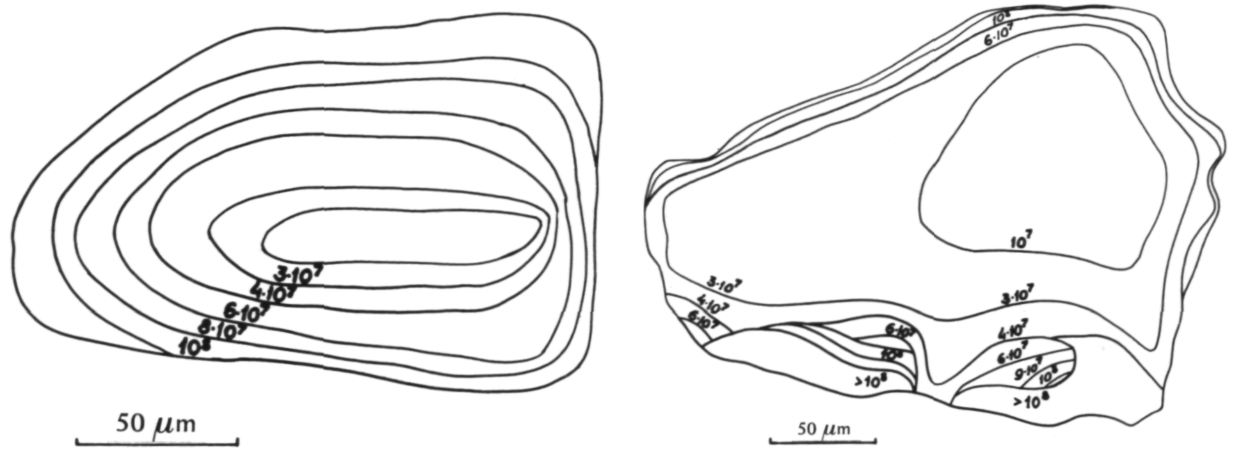


FIGURE 4.—*Track density distribution (in cm^{-2}) as a function of depth in crystals that have undergone irradiation from all sides.*

Luna 20 regolith core samples reveals the following features:

1. Among the samples from the Luna 16 regolith core, crystals with $\rho_{\mathrm{opt}} \lesssim 10^6$ cm^{-2} are not encountered, while the fraction of such crystals in the Luna 20 regolith core is about 10 percent.

2. In the track density range less than 10^7 cm^{-2}, the number of Luna 16 and Luna 20 crystals on the average is 5 percent and 35 percent, respectively, over the entire depth of the core sample, which also indicates a significant difference in the degree of their irradiation.

3. The relative fraction of strongly irradiated crystals with track density $\rho_{\mathrm{opt}} \gtrsim 10^7$ cm^{-2} is 95 percent and 65 percent for the Luna 16 and Luna 20 regolith cores, respectively.

Table 1.—*Observed Distribution of Crystals From Different Zones of the Luna 16 and Luna 20 Regolith Cores by Track Density From vH Group Nuclei of Cosmic Rays*

Regolith Site	Depth Zone in the Core (cm) (1)	Average Number of Crystals Studied	The Fraction (%) of crystals With Track Density ρ_{opt}cm^{-2}		
			$< 10^6$	10^6–10^7	$\gtrsim 10^7$
1	2	3	4	5	6
Luna 16	A (0–7)	50	—	4	96
	B (7–15)	38	—	—	100
	C (15–28)	31	—	7	93
	D (28–33)	41	—	10	90
	Average	160	—	5	95
Luna 20	I (0–5)	69	7	20	73
	II (5–10)	53	6	29	65
	III (10–15)	116	15	25	60
	IV (15–20)	56	12	27	61
	Average	294	10	25	65

NOTE: (1) Subdivision of the Luna 16 regolith core is according to Vinogradov (ref. 1). The Luna 20 core was subdivided into zones of equal length (about 20 cm), beginning with upper zone I.

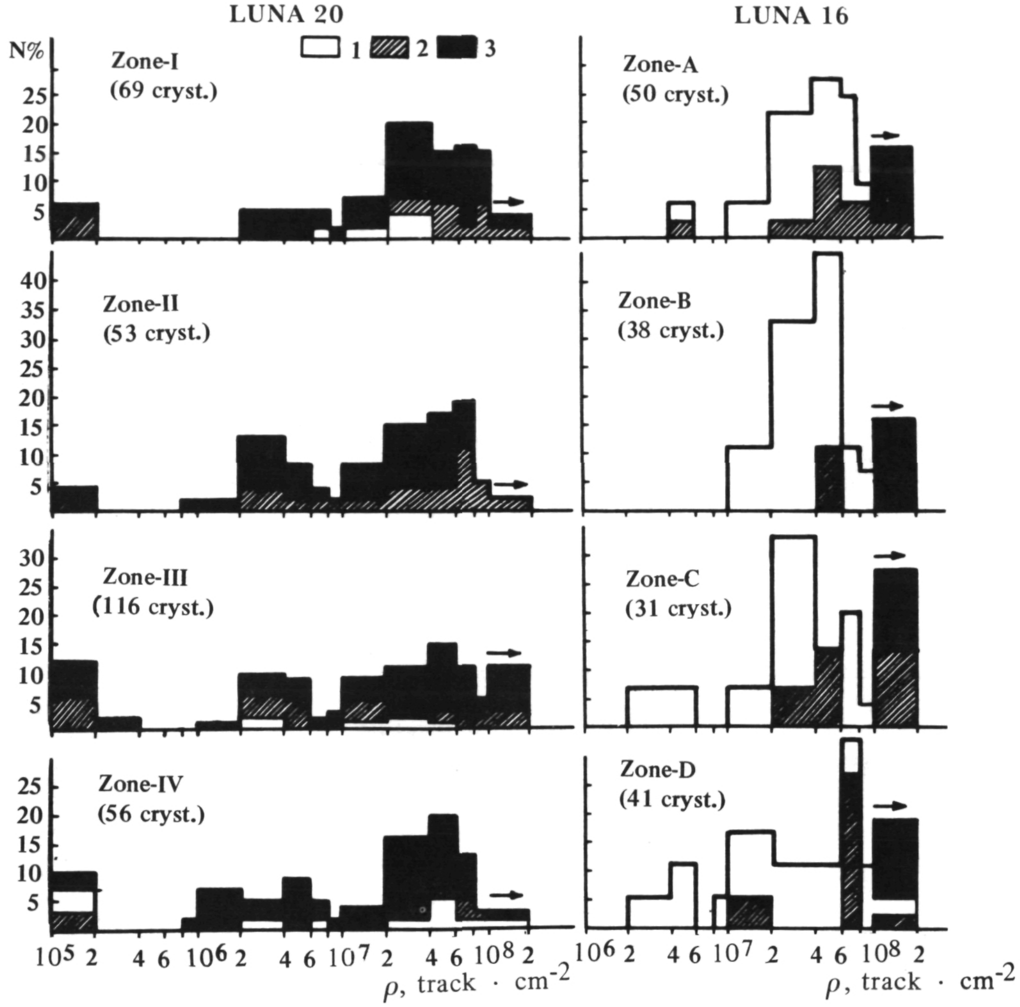


Figure 5.—*Track density distribution histograms for feldspar, olivine, and pyroxene crystals, from various zones of the Luna 16 and Luna 20 cores: 1—olivine; 2—pyroxene; and 3—feldspar.*

It also can be noted that the relative number of least irradiated samples studied, from both the Luna 16 and Luna 20 station regolith core samples (see table 1), approximately doubles for the deeper zones. At the same time, there is no depth dependence in the distribution of crystals having a high track density.

The question was examined as to whether or not the observed differences in the track density distribution of the crystals might be a consequence of differences in the mineralogical composition of the Luna 16 and Luna 20 regolith cores. First, as was pointed out above, the track density in feldspar (and pyroxene) is approximately double the track density recorded by olivine under the same irradiation conditions. Correction to the observed track density for the difference in effectiveness (see table 1, column 5), leads only to a decrease in the percentage of slightly irradiated crystals in the Luna 16 core sample. Second, the average particle size distribution of the Luna 16 and Luna 20 core samples is approximately the same (refs. 1 and 2). Therefore, the observed differences in the distribution of crystals according to their track density should not be connected with possible differential erosion effects on particles of different mineralogical composition.

For a quantitative comparison of the average degree of irradiation of the Luna 16 and Luna 20 lunar regolith material by cosmic rays, as well as for comparison with the results of investigations of other lunar samples, we calculated two parameters by analogy with Arrhenius et al (ref. 4) : N_H/N, which characterizes the fraction of crystals irradiated by low-energy nuclei of the vH group of solar cosmic rays during their passage through the upper layer of the regolith, and ρ_q, the track density corresponding to the distribution of all samples studied, of which 25 percent have a track density below ρ_q. The quantity N_H/N, calculated on the basis of our results for crystals from the Luna 16 and Luna 20 cores, gave 0.45 and 0.37, respectively. Crystals with $\rho_{\text{sem}} \overset{\sim}{>} 10^8$ cm^{-2} were used in calculation of N_H. It is evident from the data presented that only $\sim 1/3$ of all the crystals studied from the Luna 20 core sample have traces of the action of low-energy solar cosmic rays. Among the Luna 16 samples we studied, about one-half of the crystals have $\rho_{\text{sem}} \overset{\sim}{>} 10^8$ cm^{-2}. The track densities ρ_q of various zones of the Luna 16 and Luna 20 cores are between 1 to 3×10^7 cm^{-2} and 0.4 to 2×10^7 cm^{-2}, respectively. Comparison of the results obtained with other results (refs. 4, 15, and 16) indicates that the Luna 20 regolith is among the least irradiated samples of material returned from the lunar surface. One can assume that the regolith at the Luna 20 landing site, compared with the regolith returned by Luna 16, consists mainly (about 65 percent) of material, similar in degree of irradiation to the Luna 16 regolith material, and has a considerable (about 35 percent) fraction of an added component with a lower track density.

EXPOSURE AGE OF THE REGOLITH IN THE SURFACE LAYER

The rate of accumulation of tracks (ρ) from the vH group nuclei of solar and galactic cosmic rays changes as a function of the depth (X) of the sample in the regolith layer. Nuclei of the vH group of solar cosmic rays are almost completely absorbed in the upper layer $X \overset{\sim}{<} 1$mm; and in agreement with the data of Arrhenius (ref. 4), at a depth of 0.1 mm a track density of over 10^7 cm^{-2} is accumulated in 1 million years. Heavy nuclei of galactic cosmic rays with energies $E_{\text{kin}} \overset{\sim}{>} 100$ MeV/nucleon penetrate to a greater depth and, for $X \simeq 10$ cm, a track density $\rho \overset{\sim}{<} 10^4$ cm^{-2} accumulates in the same time (ref. 17). On the basis of this ratio of track density, due to heavy nuclei of solar and galactic rays, it becomes possible to estimate the effective time of track accumulation from galactic cosmic rays by using the minimum observed track density in the crystals. For example, about 10 percent of the crystals separated from zone D of the Luna 16 regolith core have a track density of not over 10^7 cm^{-2}. An estimate of the radiation age of these samples by galactic cosmic rays gives a value of $\simeq 450 \times 10^6$ years in this case, which agrees well with the results of Lavrukhrina et al. (ref. 18). The lowest track density ($\rho \overset{\sim}{<} 10^6$ cm^{-2}) obtained for samples from the Luna 20 core corresponds to an accumulation time $T_{\text{gal}} \overset{\sim}{<} 20$ million years, if the samples were under a regolith layer more than 10 cm deep. This age can be assumed as the upper limit of the time since the event that added to the Luna 20 regolith about 10 percent of material which had been practically unexposed to cosmic rays.

In the Luna 16 and Luna 20 core samples studied, crystals were encountered that contained a track density exceeding 10^9 to 10^{10} cm^{-2} at a depth of about 100μm. On the average, such crystals constituted 5 to 7 percent of both core samples. For accumulation of such a high track density from the vH group nuclei of solar cosmic rays at the present intensity and for crystals located in a surface layer ~ 1 mm thick, a time in excess of tens of millions of years is necessary. Observation of high track densities in microcrystals of lunar dust also was reported earlier by Price et al. (ref. 14). This confirms the idea of Gold (ref. 19) that microcrystals of lunar soil with very high track densities can perhaps be attributed to material which was in the free state in space before falling to the surface of the Moon.

Lunar dust particles a few microns in size from the Luna 16 core which were studied earlier (ref. 6) using a high-voltage electron microscope and without preliminary chemical etching, almost all exhibited a track density of over 10^{10} cm^{-2}. These tracks, observed in layers of micron thickness, may be entirely the result of low-energy vH group nuclei of solar cosmic rays. Consequently, the majority of microcrystals in the Luna 16 core must have been subject to such irradiation. However, the low probability of this process also leads to the suggestion that the strongly irradiated material of the lunar regolith is possibly a component that has been added to the lunar surface material from outside.

Conclusions

The following conclusions can be drawn from the data obtained.

1. The track density distributions of crystals from the Luna 16 and Luna 20 cores differ in that a comparatively large amount of less irradiated (with $\rho_{\text{opt}} \lesssim 10^7$ cm^{-2}) material is present in the Luna 20 sample, and a considerable excess of highly irradiated material (with $\rho_{\text{opt}} \gtrsim 10^7$ cm^{-2}) is present in the Luna 16 sample. This characterizes the regolith material in the Luna 16 landing area (Mare Fecunditatis) as having undergone a stronger degree of cosmic irradiation during its exposure age of ~ 500 million years (ref. 18) than the highland material returned by Luna 20.

2. About 10 percent of the Luna 20 core material is weakly irradiated by heavy nuclei of galactic cosmic rays with $\rho_{\text{opt}} \lesssim 10^6$ cm^{-2}. It is possible that this component was brought up from deep layers of the lunar surface during the formation of the crater Apollonius C and added to the main bulk of regolith, as was suggested by Vinogradov (ref. 2). On the basis of the fact that the observed minimum track density is attributed completely to vH group nuclei of galactic cosmic rays, the age of this event is estimated at less than ~ 20 million years.

3. Regardless of the depth of occurrence of the crystals studied in the Luna 16 and Luna 20 cores, specimens are encountered which bear traces of the action of low-energy vH group nuclei of solar cosmic rays. This indicates that formation of the upper regolith layer, in both the lunar mare and highland regions, includes sequential layering of finely fragmented crystalline material and subsequent mixing of it by micrometeorite bombardment.

4. The crystals with a very high track density (10^9 to 10^{10} cm^{-2}) can be attributed to material possibly brought to the lunar surface from space.

Acknowledgment

The authors express their thanks to A. P. Vinogradov for constant interest and attention to the work, as well as to L. L. Tarasov and A. V. Ivanov for selection of lunar samples.

References

1. VINOGRADOV, A. P., *Geokhimiya*, Vol. 3, 1971, p. 261.
2. VINOGRADOV, A. P., *Geokhimiya*, Vol. 7, 1972, p. 763.
3. FLEISCHER, R. L., P. B. PRICE, AND R. M. WALKER, *Ann. Rev. Nucl. Sci.*, Vol. 15, 1965, p. 1.
4. ARRHENIUS, G., S. LIANG, D. MACDOUGALL, L. WILKENING, N. BHANDARI, S. BHAT, D. LAL, G. RAJAGOPALAN, A. S. TAMBANE, AND V. S. VENKATAVARADAN, *Geochimica et Cosmochimica Acta*, Supplement 2, Vol. 3, 1971, p. 2583.
5. BHANDARI, N., J. N. GOSWAMI, D. LAL, D. MACDOUGALL, AND A. S. TAMHANE, *Proc. Indian Acad. Sci.*, Vol. 77, No. 1, Sec. A, 1972, p. 27.
6. BORG, ZH., D. DVURRO, K. ZHURE, L. L. KASHKAROV, AND M. MORETT, *Lunar Soil from Mare Fecunditatis*, 1974, p. 444.
7. KRISHNASWAMI, S., D. LAL, N. PRABHU, AND A. S. TAMHANE, *Science*, Vol. 174, 1971, p. 287.
8. LAL, D., A. V. MURALI, R. S. RAJAN, A. S. TAMHANE, J. C. LORIN, AND P. PELLAS, *Earth Planet. Sci. Letters*, Vol. 5, 1968, p. 111.

9. Kashkarov, L. L., A. K. Lavrukhina, L. I. Genayeva, M. K. Antoshin, and G. V. Spivak, *Rep. on COSPAR*, 1974.
10. Fleischer, R. L., P. B. Price, R. M. Walker, and M. Maurette, *J. Geophys. Res.*, Vol. 72, 1967, p. 331.
11. Lavrukhina, A. K., V. D. Gorin, L. L. Kashkarov, and G. K. Ustinova, *Izv. Acad. nauk SSSR, ser. fiz.*, Vol. 38, No. 9, 1974, p. 1806.
12. Crozas, G., U. Haack, M. Hair, M. Maurette, R. Walker, and D. Woolum, *Geochimica et Cosmochimica Acta 34*, Supplement 1, Vol. 3, 1970, p. 2051.
13. Lal, D., *Space Sci. Rev.*, Vol. 14, 1972, p. 3.
14. Price, P. B., J. H. Chan, I. D. Hutcheon, D. MacDougall, R. S. Rajan, E. K. Shirk, and J. D. Sullivan, *Geochimica et Cosmochimica Acta*, Supplement 4, Vol. 3, 1973, p. 2347.
15. Comstock, G. M., R. L. Fleischer, and H. R. Hart, *Earth Planet. Sci. Letters*, Vol. 13, 1972, p. 407.
16. Crozas, G., R. Walker, and D. Zimmerman, *Geochimica et Cosmochimica Acta 37*, 1973, p. 825.
17. Fleischer, R. L., H. R. Hart, and W. R. Giard, *Geochimica et Cosmochimica Acta*, Vol. 3, 1973, p. 2307.
18. Lavrukhina, A. K., and V. D. Gorin, *Space Research*, Vol. 13, 1973, p. 991.
19. Gold, T., *The Moon*, Vol. 7 Nos. 3 and 4, 1973, p. 293.
20. Kashkarov, L. L., M. Morett, P. Moris, and B. Vassan, *Report to VIII International Conference on Nuclear Photography and Dielectric Track Detectors*, Rumania, 1972.
21. Kashkarov, L. L., A. K. Lavrukhina, and L. I. Genayeva, *Lunar Soil from Mare Fecunditatis*, 1974, p. 241.