Ultrathin Amorphous Coatings on Lunar Dust Grains

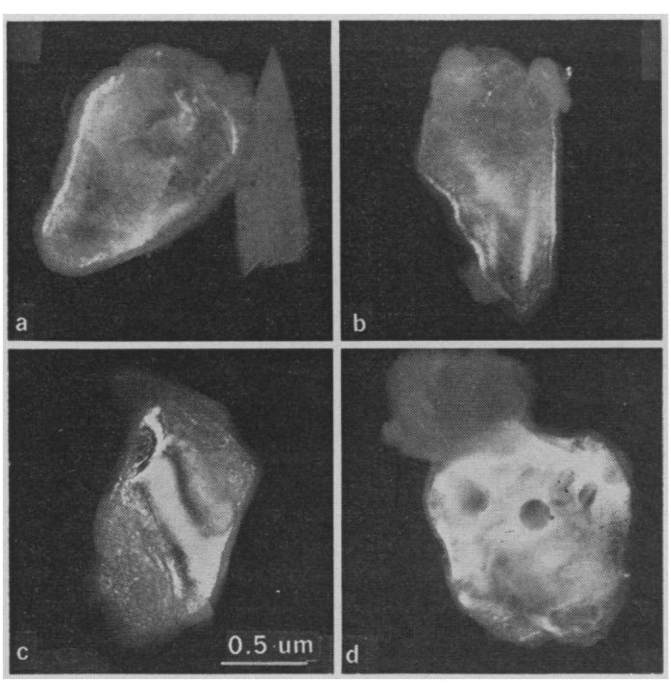
Abstract. *Ultrathin amorphous coatings have been observed by high-voltage electron microscopy on micrometer-sized dust grains from the Apollo 11, Apollo 12, Apollo 14, and Luna 16 missions. Calibration experiments show that these coatings result from an "ancient" implantation of solar wind ions in the grains. This phenomenon has interdisciplinary applications concerning the past activity of the sun, the lunar albedo, the ancient lunar atmosphere and magnetic field, the carbon content of lunar soils, and lunar dynamic processes.*

Individual lunar dust grains have been heavily bombarded by solar wind ions as evidenced by their high content of rare gases (*1*), which show elemental and isotopic abundances very similar to those measured in aluminum catcher foils deployed on the lunar surface by the astronauts (*2*). As these low-energy nuclei have very short penetrations in matter (about 1000 Å in silicates), they might produce such superficial effects in the dust grains as (i) thin "vapor-deposited" coatings resulting from solar wind sputtering (*3, 4*) and (ii) superficial metamictized layers formed by the overlapping of the radiation-damaged islands produced along the path of the incident ions (*5, 6*). Furthermore, these effects could be enhanced by α-decay products recoiling downward from a hypothetical lunar radon atmosphere (*7, 8*), with energies comparable to those expected from very heavy solar wind ions. In this report we describe evidence for the existence of metamictized layers in the lunar dust grains and summarize various implications of this new phenomenon for cosmophysics.

To search for surface coatings, we observed micrometer-sized lunar dust grains with a high-voltage electron microscope (*6*). Our main results are summarized in Fig. 1, which shows that the most rounded grains are surrounded with a dark lining of material about 500 Å thick, when observed in darkfield conditions. By using selected area electron diffraction methods, we have already demonstrated (*9*) that this dark lining represents an amorphous coating on the grains. The coating of the grains is a very general phenomenon, inasmuch as the grains in Fig. 1, a to d, were extracted from the Apollo 11, Apollo 12, Apollo 14, and Luna 16 soils, respectively.

To identify the origin of the coatings, we conducted a series of calibration experiments, the results of which are summarized in Fig. 2. (i) We crushed an internal chunk from lunar igneous rock 12021 into micrometer-sized fragments. Therefore, these fragments were never exposed on the lunar surface to low-energy solar ions, and, as expected, they have very angular habits and show no amorphous coating (Fig. 2a). We irradiated this preparation with a high dose of ^{129}Xe ions (55 kev; about 10^{16} ions per square centimeter), with the use of the heavy isotope separator at our institute (*10*), and observed the simultaneous formation in the irradiated grains of a marked rounding and an amorphous coating (Fig. 2b), similar to those observed in the natural lunar dust grains (ii). Then we studied the thermal stability of the "fossil" coatings and of the nuclear particle tracks, which were generally registered in the same grains, by heating the dust grains for 2 hours at various temperatures. We observed that the coatings and the tracks disappeared almost simultaneously at about 800°C while being transformed into small crystallites (Fig. 2c), very similar to those observed by Price and Walker (*11*) in mica artificially loaded with fission-fragment tracks and subsequently heated. We consider that this experiment shows that the coating material is similar to that constituting the radiation-damaged core of the tracks. (iii) Finally, by measuring the thickness distribution of the coatings, we determined that they were comparable to the expected implantation depth of about 500 Å, along which heavy ions of the solar wind type, with energies of about 1 kev per atomic mass unit (amu), are effective in producing radiation damage in light targets (*12*).

From these observations we deduce that the coatings are superficially metamictized layers produced during an "ancient" implantation of ions of the solar wind type in the dust grains, when they were exposed on the top surface of the lunar regolith. That these ions are most likely solar wind ions and not α-decay products of a radon atmosphere is shown by the following arguments.



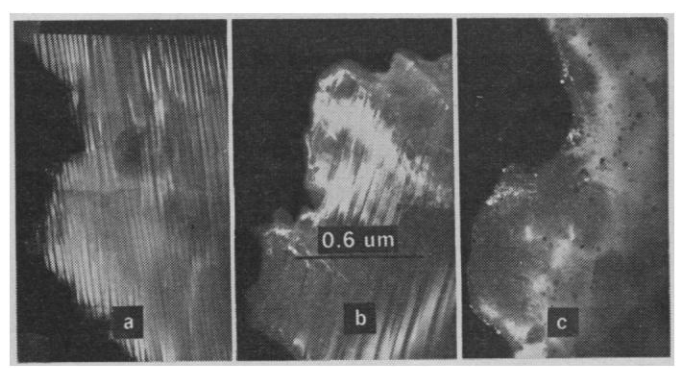


Fig. 1 (left). Darkfield micrographs (1 Mev) of micrometer-sized lunar dust grains. The grains in a, b, c, and d were returned to the earth by the Apollo 11, Apollo 12, Apollo 14, and Luna 16 missions, respectively. These grains show very rounded habits and contain a high density of nuclear particle tracks, appearing as lines of contrast. They are also coated with a very thin layer of amorphous material, which appears as a dark lining surrounding the grains. Fig. 2 (right). Darkfield micrographs (1 Mev) obtained during artificial irradiation and in annealing experiments conducted to identify the origin of the ultrathin coatings on the lunar dust grains.

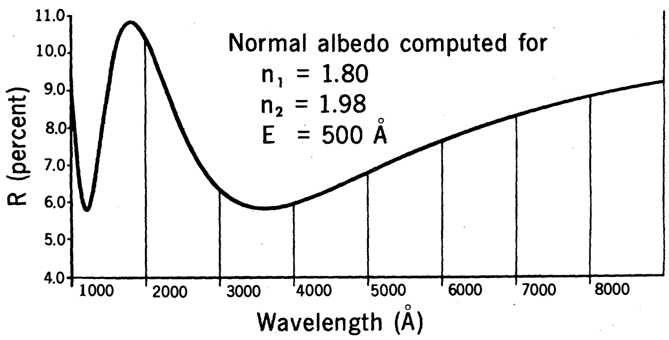


Fig. 3. Reflecting power, R, of a flat crystalline slab with an index of refraction n_2, coated with a thin layer (about 500 Å) of metamictized material with an index of refraction n_1 (less than n_2), computed as a function of the wavelength. The multidielectric structure (n_2 greater than n_1), illustrated in this figure for a slab of zircon, acts to trap incident radiation and reproduces the gross features of the spectral variations of lunar albedo, which are characterized by a smaller reduction in albedo at short and long wavelengths.

(i) Zircons are metamictized in nature by α-recoils, very similar to those produced during the α-decay of a radon atmosphere, and the minimum metamictizing doses are about 10^{13} recoils per square centimeter (*13*). We assume that such "minimum" doses are also responsible for metamictizing dust grains on the lunar surface and that the surface residence time of the grains in the radon atmosphere is about 1000 years. Then, by using the "efficient" radon diffusion model of Kraner *et al.* (*7*) and a present Th/U ratio of about 4, we infer that the concentration of U in the moon should be at least 100 times higher than the highest values so far obtained for the Apollo 14 samples. Therefore, it seems unlikely that the α-decay of a radon atmosphere is responsible for the amorphous coatings on the grains. (ii) There is a strong correlation between the proportion of coated grains and the content of rare gases from the solar wind in the lunar soil samples, but no correlation with the bulk U and Th concentrations of the soils was observed—in particular, the Apollo 14 soil contains about ten times more U and Th than the Apollo 11 soil, but its proportion of coated grains is generally smaller (*14*). (iii) Although the coating thicknesses are compatible with the effective range for radiation damage of ions of the solar wind type, the average coating thicknesses in samples 12070 and 10084 differ by a factor of 2 to 3, and that cannot easily be accounted for by the decay products of a radon atmosphere. These ultrathin metamictized layers have a number of interdisciplinary applications.

1) The albedo of a lunar soil sample could be a function of four parameters: (i) the proportion of glass (*15*); (ii) the existence of coatings on the surface of the grains (*3, 4, 6*); (iii) the concentration of strongly absorbing ions such as those of titanium (*15*); and (iv) the mineralogical composition of the samples, as the pyroxenes are more absorbing than the feldspars (*15*). In collaboration with Eugster, we have directly measured the influence of the first two parameters as well as that of the titanium concentration; furthermore, we roughly estimated the effects of the mineralogical composition by using the Al/Si and Fe/Si ratios measured in bulk dust samples by various groups (*16*). We discovered a very clear decrease in the albedo of soil samples from the Apollo 11, Apollo 12, Apollo 14 and Luna 16 missions with an increase in their proportions of coated grains (*17*). We also computed the reflecting power of crystalline grains with an index of refraction n_2, coated with a radiation-damaged layer with an index of refraction n_1. The work of Hines and Arndt (*18*) for artificially irradiated crystals and that of Pellas (*13*) for naturally metamictized zircons shows that in these cases n_2 is larger than n_1. Figure 3 shows that such a multidielectric structure can trap incident radiation by internal reflection and that the reflecting power strongly decreases in the visible range. Therefore, the albedo of the lunar soil seems to be correlated with its history of irradiation in the solar wind, as first suggested by Gold (*3*). This is an interesting result for the remote sensing of planets without atmospheres and magnetic fields such as the moon.

2) The average coating thickness in sample 12070 is about twice as great as that in sample 10084. This could reflect a change in the effective range of the particles and therefore in their energies at the time of implantation. Then it should be possible to study the past activity of solar processes that eject solar wind nuclei into space by searching for changes in the coating thicknesses as a function of the depth in well-stratified lunar core tubes. However, artificial implantations should be performed to check that the coating thicknesses depend on the energy of the ions (*19*), and one must ascertain that the depth in a lunar core tube does give a time scale for the exposure of the grains on the surface of the regolith.

3) By assuming that the minimum coating thickness of about 200 Å in sample 10084 represents a reduction in the effective range of 1 kev/amu solar wind ions due to their slowing down in a "thin" lunar atmosphere, we obtain an upper value of 10^{-9} atm for the average atmospheric pressure during the surface exposure of the grains at the Apollo 11 landing site.

4) An upper limit of about 5×10^{21} G·cm^3 has been obtained for the average magnetic moment produced by a hypothetical dipole magnetic field, located at the center of the moon and active in the past during the solar wind irradiation of the grains. This was computed by considering that the thickest coatings, about 1000 Å in sample 12070, were induced by high-energy (about 3 kev/amu) solar wind ions, unrepelled by an effective lunar magnetic field.

5) Keller suggested (*20*) that implantation of low-energy protons in silicate grains could lead to the formation of O–H bonds in the grain lattices. Therefore, part of the indigenous carbides and CH_4 observed in the lunar dust grains could have resulted from solid state reactions between implanted solar wind ions and the constituent atoms in the grains. This hypothesis seems to be confirmed by the work of Cadogan *et al.* (*21*), which indicates that the concentrations of indigenous carbon and CH_4 in various lunar soil samples are correlated to their content of coated grains, as measured by us (*6, 14*). These results initiate a field of research into the synthesis of molecules in "cosmic" dust grains individually implanted with heavy doses of "stellar" wind ions.

6) About lunar dynamic processes, we deduce from the striking similarity between the habit rounding of natural dust grains and the rounding of artificially irradiated grains (Fig. 2b) that solar wind sputtering is the major mechanism for erosion of the grains when they have been released in the lunar dust; their habits only show the scars of

such a sputtering. However, we do not claim that solar wind erosion is the controlling mechanism acting on lunar rocks, which are most likely eroded by micrometeorite chipping of their surfaces (*22*).

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19. Artificial implantations with low-energy (*E* between 0.2 and 5 kev/amu) helium, neon, argon, krypton, and xenon ions have now been performed. They show that the thicknesses of the artificial coating, although representing a complex dynamic balance between the loss and the formation of amorphous material, roughly vary as $E^{0.5}$ and do not depend on the atomic number of the ions, when *E* is expressed in kev/amu. Therefore, it is possible to study the thermal properties of the ancient solar wind by analyzing the distribution of coating thicknesses in dust samples collected at various locations in the lunar regolith. A preliminary survey of such distribution, based on 130 different "counts," suggests that the energies of the solar wind ions are clustered around an average value, which is approximately constant over periods of about 100 to 1000 years. This distribution also reveals a surprisingly high frequency of periods dominated by the emission of low-energy ions (*E* between 0.2 and 1 kev/amu) and a steep decrease in the frequency of the high-energy periods (*E* greater than 1 kev/amu), when the energy of the solar wind increases.
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Size Frequency Distribution of Martian Craters and Relative Age of Light and Dark Terrains

Abstract. *Light and dark terrains in and around Meridiani Sinus, mapped on the imagery from Mariner 6 and Mariner 7, were found to have significantly different cumulative size frequency distributions of craters. The light terrain on a mosaic of frames 6N11, 6N13, and 6N19 has a greater proportion of large craters and a lesser proportion of smaller craters than the dark terrain on the same frames. The light terrain is interpreted to be generally older than the dark terrain. The filling or partial filling of the smaller craters on the light terrain by surface detritus is suggested. Several wide-angle frames have remarkably similar cumulative crater size frequency distributions that may be representative of a large portion of the martian surface.*

The general characteristics of the martian surface, the size frequency distribution of craters visible on some Mariner 6 and Mariner 7 frames, and various crater shapes have been reported (*1*). The distribution and crater characteristics of light and dark areas of the martian surface also have been studied (*2*). However, earlier studies revealed neither obvious correlations of crater abundances with the albedo of the surface, nor differences in crater morphologies across the contacts between light and dark areas in different localities.

We selected three Mariner 6 near-encounter images (6N11, 6N13, and 6N19) having approximately the same resolution and drew the approximate contact between light and dark (higher and lower albedo) terrains, using both the photometrically corrected and the maximum-discriminability imagery (Figs. 1 and 2). The area investigated lies within 5°N to 22°S latitude and 348° to 13°W longitude (*3*). The craters on these two different albedo surfaces were identified and measured (181 craters in dark terrain, 147 craters in light terrain), and the crater diameters were corrected for spacecraft viewing angle. Craters smaller than 5 km were not counted because of the difficulty of recognition and accurate measurement at the resolution of the images. The exact position of the light-dark contact has little influence on the cumulative size frequency distributions because only a small percentage of the craters are within the area of uncertainty of the contact. The craters identified are not entirely coincident with those measured by Cutts *et al.* (*2*) on the same frames, probably because of unavoidable subjectivity in the recognition of indistinct craters (*4*). The cumulative crater size frequency distributions were plotted with probability ordinates (Figs. 3 and 4) so that random samples from like populations or the same population would graph identically, provided that the sample size is sufficient. Differences between curves are solely a function of differences in the size distribution within the populations, and cratering density differences are nowhere displayed on these curves [for further discussion see (*5*)]. The cumulative size frequency distributions of craters on frames 6N11, 6N13, and 6N19 (Fig. 4) appear to be part of the same population. However, if the cumulative size frequency distributions of craters on the light and dark terrains of the mosaic are plotted separately (Fig. 3), a large displacement (*6*) of the two curves occurs between the 20-km and 50-km crater diameters, and possibly extends to even larger crater diameters. The divergence of the cumulative size frequency distribution curves is caused by a greater proportion of larger craters in the light terrain as compared to the dark terrain. More-