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THE SEARCH FOR THE CAUSE OF THE LOW ALBEDO OF THE MOON



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INTRODUCTION

The first detailed studies of the optical properties of the lunar surface by classical astronomical methods date back to Lyot (1929), and were continued by Dollfus (1956). Their studies showed the Moon to have very specific and unusual optical properties in relation to the albedo, the angular scattering Law and the law relating polarization to scattering angle. All these quantities were determined for many areas and for specific topographic features, and it was seen that the variation over the Moon was quite small, indicating that a similar surface covered almost all regions. Lyot and Dollfus, after making a comparison with terrestrial substances, concluded that the microscopic structure of the surface had to be rough on a scale not much larger than the wavelength of light to produce the observed unusual feature in the polarization behavior, and that it had to possess an intricate shadow-casting coarse scale structure in order to result in the enhancement of the scatter intensity in the back direction, i.e. close to zero phase angle. Lyot and Dollfus realized that a fine rock powder would serve best to give the small scale roughness, and they observed that most terrestrial rock powders of sufficiently small grain size are much more reflecting than the Moon. Some volcanic ash and some iron oxide rich basalt powders were the only commonly. occurring materials that fitted the low albedo and polarization laws. For the coarse scale structure

needed to account for the backscatter, Dollfusconsidered a vesicular rock surface coated everywhere by a thin layer of the fine, dark powder. A review of this subject was given by Hapke (1971).

On the basis of a variety of lunar observations, chiefly those demonstrating the presence of erosion, Gold proposed (1955) that the Moon was covered almost everywhere by a substantial layer of dust. While this would provide the finescale roughness desired by Dollfus, it did not provide for the possibility of a coarse-scale roughness such as the proposed vesicular lava surface. Addressing themselves to the question whether a thick layer of powder could account for all the optical properties, Hapke and Van Horn (1963) measured the light scattering law for a great number of powdered terrestrial rocks. They concluded that the strong backscatter effect observed on the Moon could indeed be understood in terms of a loosely packed fine powder, so long as this was composed predominantly of small, opaque and very dark particles. However, they also observed that most terrestrial rock powders have a much higher albedo, resulting not only in a lighter surface, but also a more diffuse scattering law, and furthermore a failure to reproduce the negative branch of the polarization versus phase angle curve so characteristic for all lunar terrains. Gold (1955) had suggested that solar ionizing radiations might darken the color of the lunar surface material and

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had considered the sputter action of the solar wind as a general weathering agent, also resulting in the preferential removal of oxygen. In an attempt to discover whether this suggestion would indeed allow some more common rock powders to match the entire range of the lunar optical properties, Hapke investigated experimentally the "weathering" effects that the lunar surface would be exposed to. A number of other investigators including Wehner, Dollfus, Kenknight, Nash and Connell, also studied the effects, particularly of simulated solar wind on the optical properties of rock powders. Again, the review by Hapke (1971) covers this phase of the investigations.

After the finding that the effects of ultraviolet light and X-rays on the albedo of most rock powders were rather small, Hapke concentrated on the investigation of proton and alpha-particle bombardment in the 2 keV energy range corresponding to the solar wind. He found indeed that all the characteristic optical properties of the lunar surface could be reproduced by exposure to a 10-100 coulombs/cm² dose of 2 keV protons or alpha-particles (the latter being a more efficient darkening agent) of a variety of finely ground-up iron bearing silicates. <u>Hapke explained this</u> <u>darkening effect by a sputtering mechanism</u>. In the initial stage of the irradiation experiments (in which he used protons) he suspected that chemical sputtering, reduction of metal oxides to free metal by the monoatomic hydrogen, might have

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played an important role in the darkening effect (Hapke and Van Horn, 1963). Later (Hapke, 1966) he came to the conclution that the optical effect was mainly caused by the selective redeposition of atoms removed from the surface by sputtering. The heavier atoms, he considered, have a higher sticking coefficient and thus are recaptured by adjacent surfaces with a greater probability than the lighter atoms. This process forms an oxygen deficient heavy atom enriched coating on the individual grains, altering the optical properties of the material. In the case of iron-bearing silicates, the surface enrichment in iron that would result would make the grains optically more absorbent and more opaque. He succeeded in demonstrating that loosely packed powders of a number of commonly occurring minerals would, after such treatment, match rather well the entire range of the lunar optical properties, and in particular that among them, basalt powder was the closest match (1966).

The availability of lunar material in 1969 immediately eliminated certain explanations for the peculiar optical behavior of the Moon, whereas others are still subject to debate and are still not settled. It became obvious at once that the lunar dust cover is indeed not an inherently dark, exotic material. Ground-up lunar rocks exhibit the same range of albedo and polarization characteristics as terrestrial rocks and are, in general, much lighter than lunar dust of similar chemical and mineral composition.

Effects of lunar weathering processes were recognized as soon as analysis of the Apollo 11 dust samples began.

The considerable amount of glass of various forms mixed in the regolith was the most obvious and was interpreted as the product of a <u>micrometeorite impact melting-</u> <u>vitrification process.</u> Adams and McCord (1971) and Nash and Connell (1971) postulated that it is the mixed in, impact produced glass, which alters the optical properties

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of the dust and renders its light absorption spectrum distinctly different from that of the ground-up rock. Adams and McCord (1971) compared the reflectance spectrum of a ground-up Apollo 12 rock sample to the spectra of the same rock powder mixed with various quantities of pulverized laboratory produced glass from the same rock. As the quantity of glass mixed in the rock powder increased the characteristic absorption bands of the rock spectrum became fainter, the spectrum flattened and the reflectivity decreased over the entire wavelength range examined. The shape of the spectrum of the rock powder mixed with the glass in the same proportion as found in the Apollo 12 dust indeed resembled that of the dust itself, but its albedo was still much higher. The authors offered several explanations for this fact, and we shall return to this point in the discussion section. Adams and McCord (1973) later. examined the relationship of reflectivity and agglutinate content in a great number of Apollo 16 soils and demonstrated that the reflectivity of these samples is a function of agglutinate content, thus soil maturity. (Soil maturity is a term used to indicate the length of time during which the soil has been exposed to external weathering agents.)

At this stage there were therefore two entirely separate processes under consideration for producing the darkening of the lunar soil, and both would be acting on the surface and thus show similarly a correlation with the

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soil maturity. The question was entirely, which produced the dominant effect. Hapke had already demonstrated that sputtering could make basalt powders as dark as required and make them match the unusual polarization curve; the agglutinate admixture proposal still lacked such a demonstration.

Hapke et al (1970) and Gold et al (1970) found evidence of opaque coatings on dust grains which they were inclined to attribute to <u>the sputtering effect of the</u> <u>solar wind</u>. Hapke also presented results of proton irradiation experiments with an Apollo 11 rock powder. The albedo of the latter decreased from 0.18 to 0.12 by a dose of 20 coulombs/cm² of 2 keV energy protons. The albedo of irradiated rock powder therefore became close to that of the soil (0.09 when measured at 6° phase angle) but not quite as dark. This point will also be discussed later.

Subsequently Hapke et al (1971) raised the further possibility that <u>the process of meteorite impact induced</u> <u>evaporation and recondensation</u> might be a major contributor in the formation of the opaque coatings on the soil grains and thus the darkening of the soil. Isotopic enrichment in Si³⁰ (also in C¹³ and O¹⁸) on soil grain-surfaces has been observed in samples from all the Apollo missions by Epstein and Taylor (1970, 1971, 1972 and 1975) indicating the existence of surface alteration processes. Fractional vaporization or fractional condensation during impact melting

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or during solar wind bombardment were mentioned by these authors as possible causes of enrichment in the heavy isotopes.

Borg et al (1971) found evidence of amorphous coatings on soil grains by high voltage electron microscopy. These coatings appeared on the 1 meV dark field micrographs of the grains and their thickness distribution was peaked at about 500 Å. The origin of the coatings was attributed to either the effect of vapor

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deposition on the grains or to the effect of solar wind in the form of sputtered coatings or of radiation damaged layers on soil grains. The optical consequences of these coatings were not determined.

The recent development of a variety of techniques for chemical analysis of solid surfaces, yielding information on the outermost few atomic layers, has permitted the study of chemical changes on the surface of soil grains caused by lunar weathering processes. A two to threefold increase in the iron/oxygen ratio on the surface of lunar soil grains compared with their bulk composition was demonstrated by Auger spectroscopy (Gold et al 1974). Vinogradov et al (1972) presented evidence for the existence of metallic iron in a finely dispersed state on the surface of Lunar 16 and Apollo 11 soil samples by ESCA (X-ray photoelectron spectroscopy). The peak belonging to metallic iron had an intensity of 10-15% of that of the oxidized iron peak. Housley and Grant (1975) compared ESCA results of the less than 20µ size fraction size of soil sample 10084 to that of the 75-150µ size fraction crushed to a similar size. The spectrum of the lunar exposed surfaces showed evidence of Fe in the Fe^o oxidation state which was completely absent in the surface exposed by crushing. The above results pointed to a process which probably both increases the iron concentration on the surface of soil grains and reduces some of the iron to the metallic state.

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The optical significance of the increased iron/oxygen ratio on the surface of lunar soil grains has been discussed by Gold et al (1974, 1975). This effect was shown to be closely related to the albedo of the soil in numerous samples from all the Apollo missions suggesting that the darkness of the soil is generally due to a thin coating on the grains which is enriched in iron. As was pointed out by Gold et al (1975) these coatings might correspond to those reported by Borg et al (1971). However neither the thickness of the chemically altered coating observed by Auger spectroscopy nor the chemical nature of the physically altered coating observed by high voltage electron microscopy have been yet determined thus the above correspondence has not been established.

Gold et al (1974) considered that the two most likely mechanisms for producing the surface iron enrichment they had observed were solar wind sputtering and meteoritic evaporation and subsequent recondensation, in accord with Hapke et al (1970, 1971). These mechanisms have been extensively discussed by Hapke et al (1975) and Cassidy and Hapke (1974) as principal darkening processes on the lunar surface and possibly on the surfaces of other airless bodies in the solar system. Hapke et al (1975) demonstrated that deposits from vapors generated by either hydrogen ion sputtering or thermal evaporation (of lunarlike material) are enriched in iron, have larger, nonselective optical absorptivities and contain abundant

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submicroscopic, super-paramagnetic grains of metallic Feall of these are characteristics of the lunar soil.

At the present we have therefore the following weathering processes discussed in the literature as candidates for the cause of the darkness of the lunar soil:

- Formation of light absorbent coatings on soil grains by the following means:
 - a. Radiation damage by the solar wind, producing amorphous coatings only crystallographically different from the host grains.
 - b. Similar to la but with the added effect that solar wind hydrogen is present and changes the chemistry of the coatings.
 - c. Solar wind sputtering chiefly causing the selective removal of some elements.
 - d. Solar wind sputtering chiefly resulting in the selective capture of sputtered atoms on other surfaces.
 - e. Deposition of chemically differentiated vapor distilled material by impact vapor
 - ization and recondensation.
- Admixture into the soil of dark agglutinates produced by impact melting. The light absorption of these particles may be due not only to the

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vitrification that tends to darken by itself, but to the impact heating facilitating the diffusion . of solar wind implanted hydrogen (Housley, 1973), which reduces some of the iron to a metallic state. In the following we will concentrate on process lc We will report our latest experimental results and ld. on Auger analysis of lunar soil and rock samples as well as the results of solar wind simulation experiments with a variety of terrestrial and lunar rock samples. Our observations show the correlation of surface chemistry and albedo, and we shall discuss the possible solar wind sputtering mechanisms which can bring about the observed chemical composition on the surface of soil grains. In the discussion section we will also compare the different weathering processes enumerated above, and attempt to evaluate their relative importance for the formation of the physical and chemical state of the surface cover and for the low albedo.

EXPERIMENTAL METHODS AND OBSERVATIONS

Our recent experimental work concerning lunar weathering and its effect on the albedo of the surface cover has consisted of:

 Determination of the surface chemical composition of lunar soil and ground-up rock samples by Auger electron spectroscopy.

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- Measurement of the optical albedo of these samples.
- Proton or α-particle irradiation of terrestrial rock chips and rock powders and of ground-up lunar rock samples in order to determine the optical and surface chemical effect of simulated solar wind.

The Auger spectrometer used in these experiments was built in our laboratory and was described in previous publications (Gold et al., 1974, 75), along with the method of sample preparation and positioning in the spectrometer. The samples of lunar fines were analyzed in the same state as received from the curator (air exposed); the rock samples were pulverized in a boron carbide mortar to approximately the same mean particle size as the fines. All our results presented here were obtained with a 2000-V primary electron energy (unless otherwise stated) and with the primary beam current between 0.5 and luA. . The Auger analysis methods we developed overcame the difficulties of charge build-up on insulating powders such as the lunar soil. We considered it essential to investigate surfaces of bulk soil samples, rather than individual selected grains, to obtain information about the average surface properties of the grains, just as in the determination of the optical albedo.

The apparatus used for the solar-wind simulation experiments was essentially the same as the one Hapke had used earlier (with the addition of a forepump molecular sieve trap) and as described by him recently (Hapke, 1973).

In these experiments we bombarded lunar rock powders with protons or α particles accelerated to an energy of 2.0 keV[±] 5%. The current density of the ion beam, as measured with a movable Faraday cup, was 0.15 mA/cm² and its diameter about 10 cm. Other experimental details were described previously (Gold et al, 1975).

The experimental procedure for the solar-wind simulation experiments was as follows. Powder samples were prepared first for Auger analysis on copper sample holders. Auger spectra were taken of the samples (at least 3-5 spectra were recorded at various locations on the surface). The samples were then transferred to the ion irradiation chamber which was evacuated to base pressure (usually overnight). The irradiation period which followed was 5-7 hr (except in one experiment, where the irradiation period was 56 hours and the irradiation dose approximately 30 coulomb/cm²) resulting in 2.7-3.8 coulomb/cm² doses. After irradiation dry N₂ was admitted in the chamber and the samples were carefully transferred again to the Auger spectrometer for analysis of the irradiated surfaces.

The albedo of the irradiated samples was measured only in one case: after the 56 hour α -particle irradiation experiment. In the other cases visual darkening was observed but the albedo was not measured.

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Observations Concerning the Surface Chemistry and Albedo

We examined with Auger spectroscopy a total of thirteen samples of lunar fines (from all six Apollo missions) and six pulverized rock samples (from Apollo 11, 14, 15, and 16). The bulk chemical compositions of all these samples are available in the literature and are presented in Tables la and lb. With the retarding grid analyzer used to obtain our Auger spectra, detection of low-energy peaks is difficult. Thus of the major elements present in lunar material we observe clearly distinguishable peaks due to oxygen, calcium, titanium, and iron only. (Recently the retarding grid analyzer was replaced by a cylindrical mirror analyzer, which permits us to observe the silicon peak as well. We shall publish the related results shortly). We measured the peak height of the 650-eV iron peak, the 387-eV titanium peak (in Ti-rich samples) and the 290-eV calcium peak and tabulated the peak-to-peak height ratios of each of the above peaks and the 510-eV oxygen peak. Due to the similar chemical structure of the lunar samples examined the peak height ratios are good approximations of the surface chemical concentrations in these samples. The Auger observations were calibrated by the bulk chemical concentration reported for the rock samples, specifically by the bulk iron and calcium content of rock 60017 and by the bulk titanium content of rock 10057. We used the

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following calibration formula, for iron, for example:

1	Fe	peak	height\	measured	in	Gamplo	7							
ĺ	. 0	peak	height	Incub ut cu		Sampte	л х	bulk	न्य	atomic	concentration	יי. ז'רי	ເດັ່ງ ເປັນ	_
1	Fe	peak	height	measured	in	rock	•	. Duin	ц	aconice	concentration	111	00017	-
	0	peak	height/			600J	17				,			

= surface Fe concentration in sample A.

This formula assumes that (1) there is no difference in bulk and surface (Auger) concentration in the case of a freshly pulverized rock sample and (2) the oxygen content in all the samples is approximately the same (see Tables la and lb) and it is unchanged on the surface of soil samples. The second assumption is of course not valid if one considers that the most plausible change which occurs on the surface is a loss of oxygen. Accordingly the observed two- to threefold increase in Fe/O peak height ratio in soil samples versus rock samples might be due to a two- to threefold decrease in oxygen content. However, the above formula is still not much in error, so long as the other light elements are also depleted in approximately the same ratio. (A strong indication that this is so is the unchanged Ca/O peak height ratio and thus approximate Ca concentration in soil relative to rock samples.) To illustrate this point, let us consider the following typical bulk atomic composition for a soil ' sample: Fe: 5%, O: 60%, others (mostly Ca, Mg, Al, and Si): . 35%. If under lunar weathering one-half of the oxygen atoms leave the surface, along with one-half of the

light elements, the surface will have for every 5 atoms of iron only 30 atoms of oxygen and 18 atoms of light elements. The surface iron concentration will be $\frac{5}{53}$ ~ 10%. Thus a twofold increase in the Fe/O peak height ratio indeed closely corresponds to a twofold increase in iron concentration even if it is due to a removal of oxygen (and other light elements). It makes very little difference which rock sample is used for normalization, since the Auger concentration of the elements in question show very good proportionality with the bulk concentration of these elements in the rock samples. Only our titanium determinations caused problems, due to the generally lowtitanium content (and thus low signal-to-noise ratio) in even the "Ti rich" samples (and also due to a possible Ti contamination by the ion pump of the Auger vacuum system).

Table 2 compares the bulk iron to surface (Auger) iron concentration in our samples. The iron concentration on the surface of the soil samples is two to three times greater than in the bulk material, the average surface-tobulk iron concentration ratio being 2.4.

Table 3 compares the bulk titanium to surface (Auger) titanium concentration in samples with a significant Ti content. Although (as mentioned above) the Ti data are somewhat less reliable than the iron concentrations we determined, a tendency for increased Ti concentrations.

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on the surface of soil samples seems clear. The average surface-to-bulk titanium concentration ratio is 1.9 for the soils.

Table 4 lists the bulk and surface calcium concentrations. There seem to be no significant changes in calcium concentration on the surface of the soil samples.

Figure 1 illustrates the bulk iron + titanium, the surface (Auger) iron + titanium concentration and the optical absorption for all samples so measured. The absorption is defined as 100%-albedo. The albedo was measured at $\lambda = 5500$ Å with a narrow (230 Å) bandwidth filter at 8° illumination angle, 0° observation angle, and was normalized to an MgO standard. The crushed rock samples are light, high-albedo powders with the exception of the pulverized basalts of the Apollo 11 site. These samples all contain 2.5 - 3.5 atomic percent titanium (versus 0.1 - 0.7% Ti in the other rock samples) and it is probable that the very dark ilmenite phase in these rocks is at least in part responsible for their relatively low albedo.

In Figure 2 the albedo of ground-up rock samples and that of soil samples is plotted against the surface (Auger) iron + titanium concentration and also against the bulk iron + titanium concentration in the samples. The data points (with the exception of that of the iron poor, very high albedo soil samples 73241, 67601, 63501) are fitted to the exponential law: $A + A_o e^{-n\sigma}$ where A is the observed albedo, A_o is the

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hypothetical albedo at n = o (the law does not seem to be valid for soil samples at very low n values), n is the iron + titanium concentration (surface or bulk) observed and σ is the absorption coefficient. There are three distinct curves.

- 1. Albedo of soil samples vs. bulk iron,
 - + titanium concentration.
- Albedo of soil samples vs. <u>surface</u> iron + titanium concentration.
- 3. Albedo of ground-up rock samples vs. surface (approximately same as bulk) iron + titanium concentration.

Results of the Solar-Wind Simulation Experiments.

The Auger analysis — proton or α-particle irradiation. — Auger analysis experimental sequence was performed on olivine basalt rock chips and on several pulverized lunar rock samples.

Table 5 lists the changes observed in the surface concentration of various elements due to proton or α -particle bombardment of lunar samples 14310 and 15556 along with the mode and dose of irradiation. As can be seen from Table 5, a considerable increase of the iron to oxygen ratio was observed in each experiment when the directly bombarded grain surfaces were analyzed i.e. the Auger primary electron beam-ion beam angle was 0°-60°. The calcium/oxygen ratio changed much less markedly. We are in the process of performing irradiation experiments on titanium rich rock samples, in order to observe the change in the surface concentration of Ti. The cylindrical mirror analyzer mentioned earlier will enable us to analyze the silicon concentration and thus the irradiation effect on this element as well. Table 5 also shows that the Fe/O ratio increased much more on the directly irradiated grain surfaces than on the underside of the grains, where sputter deposits might accumulate. (Information on the underside of the grain was obtained with 120° Auger. primary electron beam-ion beam angle.)

In Figure 3 we see the Auger spectrum of the pulverized 14310 rock sample along with the spectrum of the same sample obtained after a 3.2-coulombs/cm² dose of proton irradiation (corresponding to an approximately 3000-yr. dose of the proton component of the solar wind, assuming a proton flux on the Moon of 2×10^8 protons/sec/cm²). For comparison the Auger spectrum of soil sample 14163 is also shown. The latter was collected from the same site as the rock sample and its bulk chemical composition is also very similar to that of the rock (Tables la and lb). It is evident from this figure that the equivalent of a relatively short exposure to solar wind changes the surface chemistry of the rock to that of the soil. In particular, the iron/oxygen ratio on the surface of the rock increased to a value which is within experimental uncertainty the same as that found on the surface of the soil.

We also performed a proton irradiation experiment where the targets were two olivine basalt rock chips placed one

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on the top of the other in such a way that sputtered-off material from the lower chip could be collected on the underside of the top chip. The Auger spectrum of the top side of the chip, which was directly in the ion beam, showed an approximately twofold increase in the iron--oxygen ratio versus only a slight increase in this ratio observed on the underside of the rock where sputtered off material could be collected.

The above results seem to indicate that an irradiation dose of 2.7-3.8 coulomb/cm² of 2-keV protons or α -particles is sufficient to change the chemistry of the surface of lunar rock powders (at least to a depth sampled by Auger analysis) to resemble the composition of the lunar soil surface. Alpha particles were reported to be more efficient sputtering agents (Hapke, 1966; Maurette and Price, 1975), but apparently both our proton and α -particle doses were larger than the dose necessary to establish an equilibrium composition on the surface, thus our results are similar for both ions.

The albedo of the samples was not measured in these experiments because a simple visual comparison of the freshly ground rock samples, the irradiated ground rock samples and soil samples of the same bulk chemical composition as the rock indicated that darkening occurred, but irradiation doses of 2.7-3.8 coulombs/cm² were clearly insufficient to lower the ground rock albedo to the albedo of the soil.

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We performed one experiment with sample 14310 using a large dose of α -particles. Both the Auger spectrum <u>and</u> the albedo of the sample was determined before and after irradiation with a 30 coulombs/cm² dose. The Fe/O ratio increased from 0.017 to 0.03 in this experiment, thus to the same value as the one obtained with the small irradiation dose (Table 5). The albedo changed from 0.36 to 0.07. The albedo of our Apollo 14 soil samples ranged from 0.10-0.13. The 30 coulombs/cm² irradiation dose thus darkened the rock powder to a somewhat lower albedo than that of lunar soil of similar bulk chemical composition.

DISCUSSION

On the basis of our experimental results reported above__ certain conclusions can be made concerning the following: Chemical composition

Analysis of the outermost 10-20 A° layer on lunar soil and ground-up rock grains shows that both Fe and Ti concentrations (relative to oxygen) are increased on the soil surfaces compared with the rock and bulk soil concentrations. The Fe concentration is increased on the surface of most soil samples somewhat more than that of Ti. Among the other major elements the Ca concentration is not measurably increased, neither is that of Si (according to preliminary results with the cylindrical mirror analyzer). No data is available for Al and Mg. Both the surface Fe and Ti concentrations are proportional to the bulk concentrations in these elements and seem to have reached an equilibrium value of about 2 1/2 times the bulk concentration in the case of Fe and about 2 times the bulk concentration in the case of Ti.

We have not as yet measured the thickness of the outer skin on soil grains enriched in the above heavy metals. Grant et al (1974) reported that the observed iron peaks in the Auger spectrum of a single grain of an Apollo 17 soil sample disappeared after a short period of argon sputtering, indicating that in this particular case the coating was only a few angstroms thick thus presumably optically insignificant. This was, however, only one

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experiment with one soil grain, therefore statistically not significant. In order to clearly demonstrate how this heavy metal enriched skin on soil grains might alter the optical properties of the soil we definitely need to determine its thickness. Working with single grains is tedious and it is very time consuming to obtain statistically significant results. We are currently investigating methods which would permit us to estimate the average skin thickness on grains in a given soil sample. Such a method might involve secondary ion emission spectroscopy or a variation on the conventional argon sputtering Auger spectroscopy sequence or eventually both the above techniques.

Albedo

In Figure 2 the albedo and concentration data were fitted to the exponential law $A = A_o e^{-n\sigma}$ supposing that iron and titanium provide absorption centers. As we have seen the albedo of soil <u>and</u> of ground-up rock samples is clearly correlated with both the bulk iron + titanium concentration and the surface iron + titanium concentration in these samples. (Of course in the case of ground-up rock samples the bulk and surface concentrations are the same within experimental error.) The three distinct curves, 1, 2 and 3 show that:

a. The albedo of the soil samples is approximately three times lower than that of ground-up rock samples having the same bulk iron + titanium concentration,

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see curves 1 and 3. Concurrently the soil samples have higher (2-3 times in most cases) iron + titanium concentration on their surface than the bulk concentration in these elements, see curves 1 and 2. The soil must have suffered a treatment that affected <u>both</u> its albedo and its surface iron and titanium concentration, the albedo therefore seems to be intimately related to the surface chemistry.

There is a different relationship between the b. albedo of the soil samples and their surface chemistry from that existing between the albedo of the ground-up rock samples and their surface chemistry, see curves 2 and 3. A different mechanism must therefore be responsible for light absorption on the surface of soil samples and on the surface of freshly ground rock powders. In the case of soil samples it is possible that the albedo - surface iron + titanium concentration correlation means that there exists a layer on most grains, which is thick enough to be optically significant, and in which a similarly increased concentration of heavy metals exists as in the outermost few angstrom layer analyzed by Auger spectroscopy. The ESCA results mentioned earlier suggest that at least some of the iron and titanium in this outer layer is reduced to a lower oxidation state than the state in the bulk. (For iron this is the metallic state.) Absorption centers might have been created by alteration of the

chemical state of a surface layer. It is also possible that the chemically altered skin on soil grains is too thin to be optically significant and the increased light absorption in soil samples is due to a crystallographic change in the lattice. This change then seemingly goes in step with a chemical change on the outer surface and is very probably due to the same surface weathering agent. This crystallographic change, also would more effectively increase light absorption in iron (+titanium) rich, intrinsically darker grains than in iron poor, light rock powders.

Weathering Processes

Formation of light absorbent coatings.

Solar wind sputtering is a likely process for producing a chemically altered skin on soil grains (process lc and ld) radiation damage by the solar wind creates amorphous coatings crystallographically different from the host grains (process la). Our solar wind simulation experiments followed up Hapke's earlier work in this laboratory (Hapke 1966, 1973) and were designed to investigate the surface chemical effect of the solar wind (process lc and ld) and its relation to the optical effect. Hapke had already demonstrated in detail how rock powders darkened to match the albedo and other optical properties of the lunar surface due to proton and α -particle irradiation. Our experimental results with a number of actual lunar samples show that

freshly ground-up rock samples darken and the Fe/O ratio increase simultaneously (we have no data yet for Ti/O) on the surface. The surface iron enrichment seems to reach a limiting value after an irradiation dose of approximately 3 C/cm² of 2 keV energy proton or α -particles and our results so far do not indicate with reasonable certainty whether there is a dependence of enrichment per irradiation dose on the nature of the bombarding ion. Most of our irradiation experiments were performed with the ground-up . rock sample 14310; the limiting enrichment in this case resulted in a 2.5 fold increase in the Fe/O ratio. The Fe/O ratio measured on the surface of the irradiated 14310 samples is within experimental error the same as the Fe/O ratio measured on the surface of the Apollo 14 soil sample 14163 (see Figure 3). It is also interesting that the surface vs. bulk enrichment of iron in the Apollo 14 soil samples is similar to the surface enrichment in the groundup Apollo 14 rock samples after irradiation.

The albedo in these experiments decreased only slightly by the ion dose causing the limiting chemical change, and an approximate dose of 30 C/cm² of 2 keV α -particles was needed to darken the ground-up rock to the albedo of the soil. (The actual necessary dose is probably less, since our samples became darker than the soil.) At this stage we suppose that the much higher dose necessary to darken the rock to lunar soil albedo corresponds to the dose

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needed to alter the chemistry down to a thick enough layer on the grains to cause the required optical effect.

It is important to note here that a 30 C/cm^2 ion dose corresponds to approximately 30,000 years of solar wind on the Moon taking present day fluxes. Maurette and Price (1975) report that an equilibrium thickness of the amorphous coating (presumably formed by radiation damage), observed by high voltage electron microscopy, builds up in a 2000 year equivalent of solar wind bombardment. This would indicate that process la is not responsible to any great extent for the darkening of the soil. Also, according to the above authors He ions with energies between 0.2 and 3 keV/a.m.u. are a 100 times as effective in rounding and coating grains than protons. Although in our experiments the α -particles and protons were accelerated to the same energies, therefore to different energies/a.m.u. (the latter was four times less in the case of helium than in the case of protons) the fact that so far we have seen no conclusive evidence for He ions to be considerably more efficient darkening agents than protons again indicates that process la is not likely to be the principal darkening However, this latter process is likely to occur process. on a different time scale simultaneously with processes lc and ld.

Auger analysis of lunar soils and ion irradiated rock samples seem to indicate therefore that solar wind sputtering must play a major role in determining the surface chemical composition and in view of the above also in determining the optical properties of the soil.

Mechanism of the formation of absorbent coatings: distinction between processes 1c and 1d.

We propose that the soil grains we have sampled are enriched in heavy metals, principally through the action of the solar wind by the following mechanism: protons or α -particles sputter-off atoms from the surface with an efficiency which is dependent on the mass, as well as, of course, on the surface binding energies. The transferof energy from the light ions of the solar wind is more efficient to light atoms, such as oxygen, than to the heavier atoms such as iron and titanium. Under prolonged sputtering bombardment, the concentration of the different species of atoms in the outermost layers will therefore gradually change, with the heavier atoms becoming concentrated. The final equilibrium of this concentration process will be reached when the concentrations have been changed to that value at which the relative probabilities of escape of each species is proportional to its bulk abundance in the substrate. (This is the condition of detailed continuity of flow for each of the atomic species.) This implies a particular saturation enhancement of the heavy atoms on surfaces being denuded by sputtering. It does not specify, however, the thickness of the layer in which such a concentration is reached. The latter must depend on the degree to which diffusion is effective.

The destruction of crystal lattices and the energy shed by the imbedding of foreign atoms in the outermost several hundred atomic layers by the solar wind presumably favors such diffusion, and it may well be that the entire thickness of the "amorphous coatings" that have been reported by Borg et al (1971) are subject to such diffusion. The diffusion process, however, is possibly an order of magnitude slower than the build up of the amorphous coating. We presume that it is this equilibrium composition that we observe by Auger spectroscopy, at least in the darker lunar soil samples, and it shows an augmentation of the iron-tooxygen ratio of a factor between 2 and 3. With so large an augmentation it is clear that iron must be reduced to its metallic form.

The observation that in our laboratory simulations the directly bombarded grain surfaces are more enriched in iron than the underside of the grains would suggest that the above mechanism (process lc) was in this case more important than the enrichment by sputter <u>redeposition</u> (process ld) caused by a higher sticking coefficient for heavier atoms. Hapke (1966, 1973) reported the observation that the upper surfaces of rock pieces and tightly compacted powders did not darken or darkened to a much lesser degree than loosely compacted powders. He deduced from these results that it was sputter <u>deposition</u> of films on the undersides of the grains in loosely compacted powders that caused the darkening, such mechanism not being operative (or at least

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much less effective) in the case of flat surfaces with tightly compacted grains, and that the layers on the directly sputtered surfaces were optically thin. (This reasoning, however, does not take into account that loosely compacted powder surfaces might scatter light quite differently from tightly compacted powder surfaces. In the case of the loose powder there is a much higher probability for multiple scattering of the incoming light beam in which process the same amount of light is lost at . each reflection. Thus, when the soil grains are darkened, the reduction in albedo will be greater for a loose powder surface in proportion to the occurrence of multiple scattering in the cavities of the surface. Therefore, it is possible that the individual grains receive the same treatment when exposed to ion bombardment in a loosely compacted layer as in a tightly compacted one but the reduction of the albedo of the surface composed of the grains will be greater in case of loosely compacted grains. Also, there is recent evidence that rock surfaces are darker, and proportionally to their exposure time on the Moon, than the interior of the rock (Crozaz et al_r 1974).) The sputter deposition mechanism and chemical analysis of sputter-deposited material from lunar-like glass was also described recently by Hapke et al (1975).

Quite possibly, both types of sputtering mechanisms are at work in the lunar case. Our own observations do not allow us to be certain in which circumstance the

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sputter removal (process lc) or the deposition (process ld) has been the most important, but both appear to result in somewhat similar chemical surface changes. On an average surfaces must be more affected by the sputter removal than by deposition, since for the moon as a whole there must be a net loss by sputtering. It is therefore quite probable that selective removal rather than selective deposition is generally more important, although again different areas may be treated quite differently. Surface soil samples from locations shadowed from the solar wind by a rock may allow one to observe the distinction between the two processes.

Absorbent coating by process le.

Impact vaporization, process le, which has been investigated by Hapke and co-workers (1971 and 1975), would very likely cause a similar chemical change on the surface of soil grains to the one caused by ion sputtering. During this process the vaporized species are either intercepted directly by solid grain surfaces and thus redeposited in the close vicinity of the impact, or leave the surface provided their velocity is greater than the escape velocity, or enter ballistic trajectories that return to the surface. Those trajectories redeposit on the surface a material with a chemical composition not necessarily related to the local chemistry. The apparently strong dependence we observe of surface iron and titanium concentration on the bulk concentration of the soil in these elements suggests,

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however, that the chemistry of the surface coating is linked to the chemistry of the host grains. This observation favors a process of selective removal of light elements by sputtering, rather than one by meteoritic evaporation and recondensation unless in the latter process the probability of recondensation on nearby surfaces is greater than recondensation from ballistic trajectories. The theoretical investigation of this problem is complex and awaits to be done. In any case process le must be operative and we cannot as yet determine its importance.

Arguments against the formation of absorbent coatings.

It has been discussed that the observed low albedo and enrichment in iron and titanium may be due not to a coating on the grains, but instead to an admixture of very small particles of a different composition (Dran et al, 1975). This would seem to be a possibility, and, of course, a coating of all surfaces by a layer of very fine grains would have affected all our Auger and albedo measurements in just the same manner as a surface skin on the grains. However, there is evidence that no such abundance of very small grains exists in the lunar soil. "Rosiwal's theorem" (Criswell, 1975) states that the proportion of the <u>surface</u> area of a soil sample showing one class of particles will be the same as the proportion of that class by volume in the soil. (This theorem can be given strictly for the case that the exposed area represents a cut through the

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soil that is uncorrelated with the detailed structure of the soil: it is almost certainly very nearly true in the actual circumstances discussed here.) The measurements of size distribution of soil particles (Gold et al, 1971) show conclusively that there is no major volumetric component which would be required to affect a large surface area in the size fractions below several microns. Scanning electron microscope investigations confirm that the larger grains are not coated by another population of smaller (For such an explanation the proportion of the grains. area coated would have to be large, if the albedo is thereby to be diminished from the neighborhood of 15% - 20% of the crushed rock to the 7% of a dark soil.) Furthermore the solar wind simulation experiments clearly must have produced a coating rather than small accretional particles on grain surfaces.

Dran et al (1975) also reported that evaporated 10-Å thick Fe or Ti layers on feldspar gave at least 20 times greater Auger signals than the ones observed either in feldspar blanks from rock 15065 or in feldspars from the mature 10084 and 60601 soils. They deduced that lunar weathering did not produce Fe and Ti rich coatings observed by us. Indeed, signals from a pure Fe layer of $10-\text{\AA}$ thickness should be at least an order of magnitude greater than the signals due to a few atomic % Fe present in the rocks and soils. The maximum iron concentration determined by us was 13%, on the surface of the most

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mature soils. With adequate instrumental sensitivity, however, the two- to threefold iron concentration ratio in mature soil versus rock of similar bulk composition should be clearly detectable (though possibly the choice of the iron poor feldspar grains depressed the observable iron concentration).

Process 2.

There remains the discussion of the other darkening process listed in the introduction: admixture into the soil of glass and agglutinates produced by impact melting. As we mentioned earlier this process has been very extensively examined by Adams and McCord. We believe that there are certain observational evidences which strongly disfavor the theory that glass formation by impact melting is a major darkening process especially in the case of iron rich soils. These reach an ultimate albedo of approximately 6.5% at 5500 A; which is the albedo of the dark mare areas. Adams and McCord (1971) explained the fact that the albedo of Apollo 12 rock powder mixed with glass (made of Apollo 12 rock) in the same proportion as found in the Apollo 12 dust was much higher than that of the soil by arguing that "laboratory made, crushed glass consists wholly of chips and splinters that transmit more light than the spherical or equant grains of lunar glass which trap light by multiple internal reflections". Also these authors found that the most important difference between laboratory made glass and lunar glass particles was that the former did not

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stick to the mineral grains thus a greater surface area of light particles was exposed in the laboratory mix than in the natural mare soil. Now, the Apollo 17 mission brought us the orange soil sample which is composed almost 100% of the darkest possible glass produced on the Moon, since it contains as high a proportion of iron and titanium as any soil sample found so far. It is more than three times richer in titanium and almost 50 percent richer in iron than the Apollo 12 soil. The albedo of the orange soil is however higher than that of the glass made in the laboratory by Adams and McCord. The albedo of the orange soil is 0.076 at 5500 Å, the albedo of the laboratory glass was approximately 0.06. The albedo of ground-up Apollo 12 rock is approximately 0.18 and that of the soil is 0.076. Thus only if the Apollo 12 soil were composed of almost 100% glass of higher Fe and Ti content than its actual composition could the albedo be explained by an admixture of glass and pulverized rock. In reality of course the Apollo 12 soil only contains about 20% glass and if one mixes 0.18 albedo material with 20% of 0.076 albedo glass (and the albedo of Apollo 12 glass is most probably higher due to its low Ti content and lower Fe content than that of the orange soil) even if the glass has an excellent sticking quality one cannot lower the albedo to the vicinity of 0.076. The argument is even stronger in case of the Apollo 11 soil, which is the darkest one found so far with an albedo 15% lower than that of the

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orange soil. Clearly another surface treatment than admixture of glass is needed to reach the low albedo of the mare soil.

The relationship of reflectivity and agglutinate content in a great number of Apollo 16 soils demonstrated, according to Adams and McCord (1973), that the reflectivity of these samples is a function of agglutinate content, thus soil maturity. We agree that the agglutinate content is a good index of soil maturity, furthermore in the case of light, iron-poor soils (such as most Apollo 16 soils) glass and agglutinate content is likely to influence the albedo to a greater extent than in the case of iron rich soils. (1975)However Adams and Charette'stress the point that the soil darkens mainly by the admixture of an increasing proportion of agglutinates and that the albedo of the latter is not a function of soil maturity. We retraced the curves pre-(1975)sented by Adams and Charette of reflectivity vs. agglutinate content (soil maturity) for (a) soil samples, (b) the nonagglutinate fraction of the soil samples and (c) the agglutinates fraction of the soil samples, separating the light breccia source soils from the dark breccia source soils just as the above authors did. We show these curves together in Figure 4. It seems to us that there is a clearly visible trend of darkening on all three curves. Therefore, not only is there more agglutinates mixed in the more mature soil but both the non-agglutinate and agglutinate fractions are darker --- making the existance

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of another darkening process necessary. As an example, if we take the immature soil sample 61220 and mix its nonagglutinate fraction with agglutinates in proportion to the ratio of these in a mature soil sample its albedo would become 0.25 instead of 0.16, the albedo of a mature Apollo 16 soil. The albedo of 0.16 is reached by mixing <u>mature non-agglutinate with mature agglutinate and again</u> there must be other maturation processes than agglutinate formation which very significantly contribute to the darkening of the soil.

It is very important to also note here that research concerning the role of glass formation in influencing the optical properties of the lunar soil has been restricted (at least to our knowledge) to the examination of the spectral reflectivity of the samples as a function of glass content. Hapke, however, in his extensive study of the sixties investigated the polarization and scattering properties of a variety of materials as well. He found that in order to reproduce all the characteristic optical properties of the lunar surface, especially the very strong backscatter and the negative polarization at low phase angles, one is extremely restricted in the choice of material. Whereas iron rich rock powders (such as groundup olivine) darkened by the proper dose of 2 keV energy protons and α -particles to lunar albedo and reproduced the lunar polarization and strong backscatter, all other material failed to do so. Hapke showed (1971) that glass

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beads forward scatter light strongly and their polarization properties are different from that of the lunar surface. Therefore, it is probable that if one prepared a sample of rock powder mixed with glass according to lunar composition one would not be able to reproduce the polarization and strong backscattering properties of the soil. This experiment, however, still remains to be performed.

In summary: among the darkening processes examined there is experimental evidence only for solar wind sputtering to be able to reproduce both the albedo and the surface chemistry of the soil. All the other processes are evidently also operative, among these impact vaporization and recondensation might also produce the same surface chemistry as sputtering and thereby might darken the soil by a similar mechanism. There is, however, strong evidence that neither radiation damage on the surface of soil grains nor the impact vitrification process are ultimately responsible for the very low albedo of much of the lunar soil. It is clearly important to learn more about the mechanism of both solar wind sputtering and impact vaporization.

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			Rocks			
	<u>10044</u> ª	10057 ^b	10062 ^C	14310 ^d	15556 ^e	<u>60017^É</u>
Mg	3.5	4.4	. 4.4	4.5	4.5	1.5
Al	5.3	4.9.	5.2	5.2	4.1	13.0
Si	16.5	15 . 7	15.7	19.0	17.5	15.6
Ca	5.1	4.2	. 5.7	4.8	4.1	6.4
Ti	2.6	3.3	3.4	0.3	0.7	0.1
0	60.9	61.0	59.0	64.0	62.0	61.5
Fe	6.0	6.6	6.6	2.5	7.0	1.1
^a Engel a ^b ibid. ^C Golés a ^d Philpot	and Engel (1970) et al. (1970) tts et al. (197	72).				

eStrasheim et al. (1972).

f Laul and Schmitt (1973).

							•						
							<u>Soil</u>						
	· <u>10084^g</u>	<u>12070^h</u>	<u>14003ⁱ</u>	<u>14163</u> j	<u>15301^k</u>	<u>61500¹*</u>	. <u>62240</u> ^{m*}	<u>63501ⁿ</u>	<u>66041⁰</u>	<u>67601^P</u>	<u>732419</u> *	<u>75061^r</u>	<u>76501^s .</u>
Mg	4.7	5, 8	4.3	4.5	6.8	3.4	3.1	3.5	3.8	2.3	5.4	4.3	5.7
Al	6.1	5.8	7.7	7.6	6.5	11.4	11.5	.12.2	11.2	11.5	9.1	4.7	8.2
Si	16.5	18.0	18.9	18.7	16.2	, 16.3	16.3	15.4	15.5	15.8	16.5	14.8	16.1
Ca	4.7	4.2	4.5	3.8	4.0	6.1	6.1	6.5	5.9	6.0	5.2	3.6	4.6
Ti	2.1	.7	. 6	.5	.3	. 2	. 2	. 2	.2	.1	• 4	2.7	9
0	60.9	60.7	61.7	61,9	62.0	60.5	61.3	60.9	61.5	62.4	60.5	64.7	61.2
Fe	4.9	5.1	3.4	3.1	4.2	1.7	1.7	1.3	1.9	1:3	2.5	5.2	3.3

TABLE 1b. Bulk Chemical Composition in Atomic Percent

 ^gWanke et al. (1970).
 ⁿBrunfelt et al. (1973).

 ^hWanke et al. (1971).
 ^oLaul and Schmitt (1973).

 ⁱEhmann et al. (1972).
 ^pHaskin et al. (1973).

 ^jibid.
 ^qRhodes et al. (1974).

 ^kStrasheim et al. (1972).
 ^rBrunfelt et al. (1974).

 ^wRose et al. (1973).
 ^sRose et al. (1974).

*Literature data were not available for these very samples. Data in this table were found for 61501, 62241 and 73141 and used for 61500, 62240 and 73241 respectively.

	Sample	Bulk Fe Content in Atomic %	Surface (Auger) Fe Content in Atomic % Normalized to 60017
	(10044	6.0	6.6
	10057	6.6	7.2
rock	10062	6.6	7.9
TOCK	14310	2.5	2.1
	15556	7.0	6.8
	60017	1.1	1.1
	(10084	4.9	13.0
	12070	5.1	11.9
	14003	3.4	7.9
	14163	3.1	5.9
	15301	4.2	10.3
	61500	1.7	4.8
SOTT,	62240	1.7	3.8
	63501	1.3	4.0
	66041	1.9	5.3
	67601	1.3	4.2
-	73241	2.5	3.5
	75061	5.2.	14.4
	76501	3.3	7.1

	Sample	Bulk Ti Content in Atomic %	Surface (Auger) Ti Content in Atomic % Normalized to 10057
	(10044	2.6	.3 . 8
roak	10057	3.3	3.3
LOCK	10062	3.4	2.9
	15556	• 7	. 8
	10084	2.1	2.8
roil 4	12070	.7	1.9
, , ,	75061	2.7	3.3
	76501	.9	2.1

	Sample	Bulk Ca Content in Atomic %	Surface (Auger) Ca Content in Atomic % Normalized to 60017
	/10044	5.1	4.8
	10057	4.2	no data
	10062	5.7	4.0
rock	15556	4.1	4.2
,	14310	4.8	4.5
	60017	6.2	6.2
	(10084	4.7	4.3
	12070	4.2	4.0
	14003	4.5	4.6
	14163	3.8	4.4
	15301	4.0	4.3
	61500	6.1	5.4
soil	62240	6.1	6.0
	63501	6.5	7.6
	66041	5.9	5.2
	67601	6.0	7.1
	73241 [.]	5.2	4.6
1	75061	3.6	3.3
	76501	4.6	4.1

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Sample	Irradiating Ion	Irradiation dose (coulombs/cm ²)	Ion beam- sample surface angle	Auger primary Electron Beam - ion beam angle	Fe/O Before After Irradiation	Ti/O Before After Irradiation	Ca/O Before After Irradiation
					· .		
14310	proton	3.0	900	∼60 ⁰	0.017 0.04		0.51 0.66
81	α particles	2.7	45 ⁰	· 0°	0.017 0.03		0.51 0.59
**	proton	3.2	45 ⁰ ·	0 ⁰	0.017 0.045		0.51 0.50
67	proton	3.2	45 ⁰	120 ⁰	0.017 0.027	, 	0.51 0.56
15556	α particles	3.8	900	~60 ⁰	0.056 0.11		0.47 0.7

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This table was constructed with spectra showing negligible carbon contamination, as can be seen in Figures 3

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Figure 1. Comparison of the bulk iron 4 titanium content (obtained from the literature, see Table 1); the surface iron + titanium concentration, (calculated from the Auger Fe and 0 peak ratios on one hand; Ti and 0 peak ratios on the other and normalized with respect to the bulk iron concentration of rock sample 60017 and the bulk titanium concentration of rock sample 10057); and light absorption in ground-up rock and soil samples.

Figure 2. Albedo vs. iron + titanium concentration in ground-up rock and soil samples. The data points (with the exception of that of the iron poor, very high albedo soil samples 73241, 67601, 63501) are fitted to the exponential law: $A = A_{o}e^{-n\sigma}$, where A is the observed albedo, A_{o} is the hypothetical albedo at n = o, n is the iron + titanium concentration (surface or bulk) observed and σ is the absorption coefficient. The concentration error bars indicate the Auger concentration extremes obtained by taking spectra on various spots of the same sample, the albedo error bars refer to the lowest and highest albedo measured with different sample orientations. (Since very small (8 mm diameter) samples were used for the albedo measurements, they were repeated three times with three different sample orientations - in the same plane - under the light beam.) The albedo was measured at 5500A wavelength, at 8° illumination angle and was normalized to MgO.

Figure 3. Auger spectrum of the pulverized rock sample 14310, along with the spectrum of the same sample after being irradiated by 2 keV energy protons with a dose of 3.2 coulombs/cm². The spectrum of soil sample 14163 is also shown in the figure. The soil sample is from the same location as the rock and its bulk chemical composition is similar to that of the rock. The primary electron beam energies and beam currents are listed in the figure; the modulation voltage amplitude was 20 V peak-to-peak for all three spectra.

Figure 4. The reflectivity at 5650Å wavelength of Apollo 16 soils, of their agglutinate fraction and of their non-agglutinate fraction versus the weight percent of agglutinates. Samples are labeled with three digits only, thus 122 = 61221. This figure was constructed from figures by Adams and McCord (1975).





Figure 2



