SPECULAR REFLECTIONS AND THE NATURE OF PARTICLE SURFACE INTERACTIONS  
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Introduction  Light reflected from a particulate surface has undergone multiple transmissions and reflections between particles (1,2). Scattering by a particulate sample is a function of both bulk sample properties (compaction, particle size, etc.) and the scattering efficiency of the individual particles (3). The complexity of the solution to the equation of radiation transfer for real particulate samples has prompted the simplifying assumption that all particle surface interactions are specular reflections, which are approximated mathematically by Fresnel's equations (1). This approach contains an implicit assumption that particle surfaces are optically smooth for all wavelengths considered.

In natural surfaces, single particles may scatter light by single or multiple specular (Fresnel) reflections, or by resonant (Rayleigh) scattering, which occurs from particles or surface features close in size to the incident wavelength (2). Light which interacts with a surface by one of these two mechanisms is polarized in a manner diagnostic of the mechanism. The angular distribution of scattered light can also be used to differentiate between Fresnel and Rayleigh interactions. Specular (Fresnel) reflections are confined to directions for which the incident angle, $i$, equals the emergent angle, $e$, although rough surfaces may geometrically disperse light through specular interactions from randomly oriented surface facets. Rayleigh scattered rays, on the other hand, are dispersed nearly isotropically (3).

Data Acquisition  To investigate light interactions with particle surfaces, obsidian slabs were prepared with controlled surface roughness to approximate natural particle surfaces. The visible near IR spectra of three obsidian samples are shown in figure 1. Slab surfaces may be thought of as one end member of a series of particulate samples of increasing mean particle size. Obsidian was chosen to minimize internal crystal boundary reflections. The lack of significant absorption features in such slab spectra indicate that internally transmitted light is unimportant compared to surface interactions of radiation.

Two obsidian slabs were prepared by grinding with a) 400 grit (40µm) and b) 400 then 1200 grit (5µm) Al oxide powder. These will be called the rough and smooth slabs, respectively. Using the RELAB bidirectional reflectance spectrometer the reflectivity relative to halon between 0.35 and 1.8µm, and the polarization state of reflected light between 0.6 and 1.8µm, were measured for each of the slabs for phase angles between 15 and 90 degrees. Polarization determinations require measurements: 4 sample measurements with polarizing optics in both incident and reflected beams, and 2 measurements to determine the light source polarization bias. This arrangement allows the depolarization properties of the sample to also be measured in the crossed-polars configuration.

Results  Shown in figure 2 are reflectivity measurements of the smooth slab at 3 specular geometries. For all of these geometries, reflectivity is seen to undergo a transition from short (<.8µm) to long (>1.2µm) wavelengths. Short wavelengths exhibit low reflectance which increases with phase angle, $g$, while long wavelengths exhibit high reflectance which also increases with $g$. The transition zone between short and long wavelength behavior is seen to move toward shorter wavelengths with increasing $g$. Reflectivity measurements at a constant $i$, for several values of $e$, were also obtained. These show that at the longer wavelengths, the flux is strongly concentrated along the specular direction. As $g$ increases, all wavelengths show a preferential brightening along the specular direction.

The reflectivity of the rough slab for $i=45$deg and five $e$ positions, including the specular direction, are shown in figure 3. The long wavelength specular surge is absent and all curves are remarkably similar, varying only in relative brightness. No preferred direction of reflectivity is observed at any geometry.

Polarization of light reflected from the slabs for two selected wavelengths is shown in figures 4, 5. Specular ($i=e$) geometries are labeled S for the smooth slab and R for the rough slab. Points labeled M and N are the smooth and rough slabs, respectively, for which $i=0$ but the effective incident angle plotted is half $g$. The predicted polarizations for Rayleigh scattering and Fresnel reflections, which depend on the complex index of refraction, are also shown. The refractive indices used to generate these Fresnel curves are in general accord with those found in other volcanic glasses (4). For all measurements, the cross polarized terms were insignificant. (<5% as bright as the two non-cross polarized terms.)

Discussion  The dramatic reflectivity variation with wavelength of the smooth slab suggests that two modes of reflection are in operation between 0.35 and 1.8µm. Long wavelengths are reflected specularly from the smooth slab surface. This is confirmed by the polarization of these wavelengths at specular geometries (5). Short wavelengths, however, are more geometrically dispersed, apparently from randomly oriented facets. The polarization state of this dispersed light (M) indicates that short wavelengths have also undergone a single Fresnel reflection. The migration of the transition zone and the increase in brightness with $g$ of the short wavelengths suggest that these two modes are mixed in different proportions. This slab surface is thus optically smooth at longer wavelengths but optically rough at the shorter wavelengths.

For any given wavelength, all polarization measurements for both slabs (figures 4, 5) fall on the same Fresnel polarization curve, regardless of slab roughness or instrument geometry. The structure of the surface determines the degree of dispersion of reflected radiation.

These results indicate that single Fresnel reflection dominates the first surface interaction of light with particle surfaces in the wavelength range 0.35-1.8µm. Rayleigh scattering is apparently unimportant even when surface features comparable in size to the incident wavelength are known to exist.

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Fig. 1 Bidirectional reflectance spectra of two particulate and one cut slab (rough) sample of obsidian. \( \theta = 0, \phi = 30^\circ \) (Halon Standard)

Fig. 2 Bidirectional reflectance spectra of smooth slab at 3 specular geometries. Angles in deg. (Halon Standard)

Fig. 3 Bidirectional reflectance spectra of rough slab at 5 geometries including the specular direction. Emergent angles in deg. (Halon Standard)

Fig. 4 Predicted Rayleigh (A) and Fresnel (B, \( n=1.8, k=0.001 \)) polarizations for \( i=\phi \), and experimentally derived polarizations for smooth and rough slabs at 0.6\( \mu m \).

Fig. 5 Predicted Fresnel polarization (\( n=1.6, k=0.001 \)) for \( i=\phi \), and experimentally derived polarizations for smooth and rough slabs at 1.8\( \mu m \).