NOTES

Ion Bombardment of Interplanetary Dust

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The recent discovery of ion tracks in interplanetary dust and the increasing evidence for carbon and carburized materials in these objects are strongly suggestive that chemical processing by energetic charged-particle bombardment has occurred during the dust lifetimes. The track density gives a measure of the total ion fluence experienced by the grains. We use this information and laboratory data on the modification of icy surfaces by incident ions and electrons to discuss the likelihood that chondritic interplanetary dust particles could have been processed, by plasma bombardment, from aggregates of particles which had volatile and/or organic mantles. Such a processing would leave carbon and carburized deposits and can affect estimates of the temperature of formation of these dust grains. © 1986 Academic Press, Inc.

A fraction of the interplanetary dust particles (IDP's) collected in the stratosphere by high-flying aircraft are thought to be materials ejected from comets (e.g., Greenberg, 1983). If this is the case, then these particles can, in principle, provide useful and important information on cometary formation processes. However, these particles may be highly processed by solar energetic ion bombardment in the time interval between their ejection from the comet and their collection in the atmosphere (Strazzulla et al., 1985; Lanzerotti et al., 1978). In fact, it is problematic as to how Van der Waals-bonded aggregates of presumably icy material can both lose their icy mantles and remain sufficiently stable to survive entry into the upper atmosphere (Fechtig and Mukai, 1985). The fact that ion bombardment can both chemically alter and physically erode icy materials (Johnson et al., 1985) and, in addition, produce enhanced adhesion between dissimilar materials (Tombrello, 1983) has led to the suggestion that ions may play an important role both in determining the state of the collected material and in providing the required adhesion for the porous aggregates (Strazzulla et al., 1985; Johnson, 1985). We examine these possibilities quantitatively using recently published information on ion tracks and carbon in IDP's together with laboratory data on charged particle bombardment of surfaces.

Ion tracks have now been reported in the silicate IDP's (Bradley et al., 1984a). This observation not only confirms the ability of the interplanetary ions to penetrate these materials, it also provides a lower limit on the total ion fluence (total number of ions per unit area) received by these particles during their residence as free particles. The fluence is estimated to be equivalent to an exposure of about $10^4$ years at 1 AU, using present solar activity (Bradley et al., 1984a; Fraundorf et al., 1980). The average residence time in interplanetary space for IDP's has been uncertain depending on scenarios for the particle's arrival at the collectors or on the source rates for zodiacal light particles (Fechtig, 1983; Greenberg, 1983). The residence time deduced from the observed track densities is consistent with the lower limits of such estimates (e.g., Fechtig and Mukai, 1985).

For our purposes, if one assumes, to first order, similar energetic particle spectra (energies and species) in the various regions of space, the net particle fluence received by a grain would be similar whether the grain received the bombardment indicated by the track density prior to its accretion into the comet, during its time in the mantle of the nucleus, or on its traverse from the comet to the collector. Therefore, the subsequent discussion is broadly applicable. We are aware, of course, that the energetic particle speci-
Carbon compounds, interstitial carbon, and elemental carbon have also been reported in IDP’s (Bradley et al., 1983; 1984b). The carbon compounds have been cited as evidence of possible heterogeneous catalysis (i.e., high temperatures in the formation process). While this may indeed be one explanation, we discuss here a mechanism in which ion irradiation of solids can readily produce mixing and molecular bonding between layers of dissimilar materials. That is, energetic (hot) atoms produced along ion tracks in a material, either during electronic relaxation processes or in direct collisions, can readily react, thereby simulating high-temperature processes locally (Johnson et al., 1985; Rössler et al., 1984; Johnson and Brown, 1982).

In addition, ions and electrons are known to alter drastically carbon- and sulfur-containing materials, forming stable residues of carbides and oxides. (Such a situation is well known to experimentalists using electric discharges in vacuum systems containing pump oil.) Recently, such alterations have been demonstrated to occur also for frozen volatiles containing carbon (Cheng and Lanzerotti, 1978; Moore and Donn, 1983; Strazzulla et al., 1984, 1985; Haring et al., 1984, Lanzerotti et al., 1985) or sulfur (Melcher et al., 1982; Moore, 1984; Chrisey et al., 1985; Boring et al., 1985).

For volatiles on grains in the Solar System, this irradiation mechanism may be of equal or greater importance to understanding grain chemistry than invoking a high temperature of formation. It was invoked to explain the dark rings at Uranus by Cheng and Lanzerotti (1978) and has since been used in numerous other instances (Moore and Donn, 1983; Calcagno et al., 1985; Lanzerotti et al., 1986). In fact, the outer regions of the heliosphere, in which volatiles with vapor pressure lower than water are likely to have condensed, have been found to be replete with dark materials (Brown and Cruikshank, 1983; Cruikshank et al., 1983) and indeed cometary surfaces may have low albedos prior to the onset of sublimation (A’Hearn et al., 1981).

Previously we examined the role of solar particles for eroding possible ices on the lunar surface and on interplanetary ice grains (Lanzerotti et al., 1978; 1981; Johnson et al., 1982, 1983). These estimates required some extrapolation of the few laboratory data points available at that time. Here these erosion estimates are adjusted using the presently available compilation of data for H2O (Johnson et al., 1985) and assuming ~5% He++ in the plasma. The average change in thickness of a water-ice deposit (approximately 1 g/cm2) due to an equivalent exposure of 10⁴ years at 1 AU is ~15 μm for the 1 keV/amu solar wind particles, 0.6 μm for the (~10 keV) co-rotating solar particles, 0.006 μm per solar flare event, and negligible erosion for galactic cosmic rays. The penetration depths vary considerably, and scale inversely with the density (Johnson et al., 1983); ~0.02 μm for the 1 keV solar wind protons (Strazzulla (1985) assumed 0.5 μm for a low-density ice) and ~meters for the galactic particles (Moore and Donn, 1983). The tracks reported in IDP’s are assumed to be principally produced by high-Z ion nuclei from solar flare events. As such flare particles are not highly penetrating (~a few μm), the track-containing material was either of small dimensions or a surface material on a larger object during the exposure. Therefore, it is likely that any volatile surfaces initially on those IDP’s having tracks must also have experienced erosion by the solar wind and energetic solar and galactic particles.

Greenberg (1983) has proposed a model of cometary grains as having icy outer mantles, inner organic mantles, and iron-silicate cores (grains in upper half of Fig. 1). Icy mantles on a grain ejected from a comet can be lost due to sublimation as well as by solar parti-
cule erosion (upper portion of Fig. 1). The loss of this outer material becomes a problem, as many of the IDP's are fairy-castle type structures of the order of 10 μm, the units of which are much smaller grains, ~0.1 μm. It is only the larger pieces or aggregates which are collected at Earth orbit; solar radiation pressure and the sweeping of charged grains by the solar wind depletes the smallest grains and grains ≤ 5 μm are not efficiently collected. If the aggregates ejected from a comet are largely composed of Greenberg-type grains (~0.5 μm), the rapid sublimation of an icy mantle is likely to dissociate this aggregate which is only held together by weak forces (e.g., adhesion energies of the solid: 0.088 eV for CO, 0.27 eV for CO₂, 0.52 eV for H₂O). The sublimation rate, however, decreases very rapidly with increasing distance from the Sun. Therefore, aggregates ejected in regions of space where the sublimation loss rates are non-disruptive can maintain their structure while solar particle penetration and processing of the materials occurs. Alternatively, it is possible that for some IDP's the icy mantles were lost prior to or during ejection from a comet and that an aggregate from the cometary surface is ejected for prior to or during ejection from a comet and that an aggregate from the cometary surface is ejected for which only the organic and silicate fractions remain. We will first consider the case in which the icy mantles are present and we will comment on the latter scenario at various points. Before proceeding, however, we discuss briefly the expected chemistry and physics of ion processing of materials.

In traversing a solid, energetic ions break bonds, allowing the formation of new species (Johnson et al., 1983, 1985), and enhance the diffusion of atoms, molecules, and molecular fragments (Reimann et al., 1984). In forming new species, both more volatile species (e.g., H₂ and O₂ from H₂O, H₂ from polymers) and much less volatile species (e.g., C₂ from CO, SO₂ from SO₃, and C₄H₄ from CH₄) are produced (see, e.g., Johnson et al., 1985). As the atoms and molecules along the ion track are made mobile by the passage of the ion, the diffusion of volatiles is enhanced. This enhanced diffusion allows the segregation of the more volatile species over the full depth penetrated by the incident particle (Reimann et al., 1984). These more volatile species are brought to the surface where they are preferentially lost by sublimation or sputtering (Haff, 1977), a process which gradually leaves behind a less volatile residue. A clear example of this is the loss of H atoms (as H₂) from a condensed low-temperature methane ice as measured by Lanzerotti et al. (1985, 1986). A dramatic change in the H to C ratio (4 → 2) is achieved for MeV ions at fluences ~10¹⁰ He⁺/cm² and ~10⁴ H⁺/cm². Residues are also found for CO and CO₂ ice layers bombarded by keV ions (Haring et al., 1984; Chrisey et al., 1985). In fact, residues result when ions of all energies and types bombard such carbon-containing materials. For a group of organic-type materials Strazzulla et al. (1984) assign an effective cross section ~10⁻¹⁰ cm² for the conversion of the organic material to a material enriched in carbon by fast ions, which is consistent with the results of Lanzerotti et al. (1985) for MeV ions on CH₄ at small fluences. At larger fluences, smaller effective cross sections are appropriate, corresponding to an increased difficulty of making H₂ (Lanzerotti et al., 1986).

For the minimum 1-AU equivalent exposure estimated from the track densities (order 10⁻¹⁰⁻¹⁰⁹ ions/cm²), the total solar-particle fluence is ~10⁹ ions/cm². The fluences of 10⁷'s of keV protons penetrating more than 1 μm is ~10¹⁷ ions/cm², and the energetic electrons can add to this. Therefore, in the complete absence of sublimation, such levels of particle irradiation are more than adequate for producing blackened, highly carbonized and refractory surface layers from organic and volatile mantles. For carbon-containing ices a dark and fluffy residue is produced. KeV ion erosion of CO indicates that a residue containing ~5-10% of the initial carbon is formed (Chrisey et al., 1986). MeV light ion erosion of condensed CH₄ of thicknesses such as those discussed here indicate that most of the carbon in the sample remains as a residue of carbon and hydrocarbons (Lanzerotti et al., 1986; Foti et al., 1984). Therefore, an ice mantle at normal density and radial extent ~0.5 μm having a 50% mixture of H₂O and CO (or CO₂) will leave a thin carbonized film equivalent to a carbon layer of radial extent ~0.05-0.02 μm. The conversion of a pure organic mantle to such a residue is at least as efficient (Strazzulla et al., 1985). Therefore, a carbon deposit with a maximum thickness of the order of the penetration depth of the ions can be formed on a grain at low ambient temperatures.

In addition, since the penetrating ions produce mixing on an atomic scale, they can create bonds between mantle and core materials (Johnson, 1985). The carbon in the mantles can become bonded to the silicates and metals in the core, becoming chemically equivalent to a carburized material formed at high temperatures. Of course, in the immediate vicinity of an ion track (~0.002 μm) the "temperatures" are locally high (~ a few thousand degree Kelvin) for a very brief time (~10⁻¹¹ sec) (Johnson and Brown, 1982; Seiberling et al., 1982). At earlier times individual atoms can have significant kinetic energies allowing the possibility of highly endothermic reactions (Rössler et al., 1984). Therefore, at the interface of the ion track and the interface, the mixing of mantle and core atoms which are energized allows the production of interstitial species, the formation of new molecular species (for instance carbidies), and the production of enhanced adhesion.

In the absence of very rapid concomitant heating and sublimation, the material processes described above are relatively nondisruptive as the track widths are small compared to the scale of the grains. This is a distinct difference from thermal processes which would occur continuously over the entire grain. Mukai (1980) has suggested that incident particles can also disrupt grains due to stresses produced around the ion track. This has not been observed in the lab on the
scale proposed, perhaps because the energy conversion efficiency was overestimated in that paper. The principal disruptive effect of the ions is charging due to secondary electron ejection, as discussed by a number of authors (e.g., Grün et al., 1980; Draine and Salpeter, 1979). The amount of charging depends on the relative fluxes of the photons, plasma electrons, and ions incident on the surface and, therefore, decreases with distance from the Sun, as does sublimation. Hence, as the distance from the Sun increases, the principle effect of the energy deposited by an individual ion is to eject volatiles relatively gently from an \(-0.002-\mu m\) diameter on the surface.

As the most volatile material in the grain is brought to the surface and ejected, the radius of the aggregate can gradually change in the same way that the solid (condensed) films of initially volatile materials in the laboratory change in thickness during ion bombardment (Foti et al., 1984). During the erosion of the laboratory films the residual film gradually accumulates the less volatile and more difficult to sputter species, and the constituents available for forming volatiles are reduced (e.g., H in CH4). The result, after long-term erosion, is the formation of a sputter-resistant, fluffy residue (enriched in carbon), the thickness of which is proportional to the initial thickness of the mantle (Foti et al., 1984; Lanzerotti et al., 1986). This gradual reduction in the volatile content of the mantle allows the entire aggregate to maintain its integrity while leaving it coated with a blackened residue (lower portion of Fig. 1). Also, as noted above, the ions can assist in "welding" the core materials together by ion-enhanced adhesion which is, essentially, bond formation between surface and substrate materials caused by penetrating particles (Tombrello, 1983).

Sublimation can assist the above erosion process if it occurs at a rate comparable to the sputter rate (Mukai and Schwegm, 1981; Johnson et al., 1983). However, as stated earlier, the mantle may be lost and the aggregate will be disrupted under too rapid sublimation. Therefore, the process described here depends critically on the distance from the Sun at which a grain aggregate is ejected from a comet before decaying in orbit and collection at Earth orbit. If the H2O loss rate is the controlling sublimation process then distances \(-4\) AU are required for sputtering and sublimation to be comparable (Johnson et al., 1983; Hanner, 1981; Mukai and Schwegm, 1981). Such distances are similar to those discussed when describing the decaying orbit of an ejected grain collected near Earth orbit (e.g., Greenberg, 1983).

The Helios dust experiment observed two different dust populations: dust orbiting the Sun with low eccentricity but having high densities (3–8 g/cm³) and low-density (\(\leq2\) g/cm³) grains with higher eccentricities (Grün et al., 1980). The latter are thought to be more recent cometary ejecta. We have proposed here an erosion process which would allow compaction of IDP's in a nondisruptive manner. The decay to a less elliptical orbit could in principle proceed simultaneously and in fact requires considerable time. Fechtig and Mukai (1985) have described a different scenario for particles to lose material without break-up while decaying to less eccentric orbits. They point out that the differences in sublimation rates from the solar-facing and dark sides of a grain would produce a compaction force, allowing the aggregate to maintain its structure during sublimation. In their discussion they refer to sublimation of both icy materials and the organic mantle. Their compaction scenario assumes that temperature differences can be maintained throughout the grain and that the between-grain ices can be ejected in a nondisruptive manner, even on the closer approaches to the Sun. If this is a viable process, then particle irradiation processing of the materials can occur concomitantly. Over the range of the average erosion rates suggested (5 to \(5 \times 10^{-13}\) g/cm² sec or 1.5 to \(5 \times 10^{10}\) mol/cm² sec if H2O for a particle in a highly elliptical Halley-like orbit, they suggest it takes \(10^4 \sim 10^5\) years to lose the icy and organic mantles. This is similar to the time required for the particles to decay from the highly elliptical orbits into orbits of low eccentricity (\(-2 \sim 10^5\) years). In such time intervals we have shown, as has Strazzulla (1985), that a very fine layer of carbon-enriched material (equivalent to \(0.05 \mu m\) at normal densities) can be formed from the ices and the organic mantle material by particle processing.

In summary, the observational discovery of particle tracks in certain IDP's clearly indicates the exposure of these particles to \(\sim10^4\) years of 1-AU equivalent solar-particle fluences. If some erasure of the tracks occurs, which is likely when an IDP enters the upper atmosphere (Fraundorf et al., 1982), then somewhat longer times are implied. At such levels of charged-particle fluence the icy and/or organic mantles of grains can be chemically processed and eroded, leaving blackened deposits like those often observed on IDP's with compounds that generally form at high temperatures. Furthermore, the erosion and enhanced adhesion produced by ions can account for the fact that what are initially weakly attached aggregates (bonded by Van der Waals forces) can survive as the chemically bonded structures collected at Earth or from lunar samples. If such ion processes are important for IDP's then we would also expect the ability of the ions to alter the physical structure (e.g., creation of voids and of crystalline and amorphous regions) to be important in determining the state of the collected particles.

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