New frontiers in the Solar System: trans-Neptunian objects/Les nouvelles frontières
du système solaire : les objets transneptuniens

Ion irradiation of TNOs: from the fluxes measured in space to the laboratory experiments

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Abstract

Cosmic ion irradiation is believed to be one of the processes driving the evolution of the surface materials on TNOs. We review the laboratory simulations of radiation effects induced in likely TNO materials. In particular the production of new molecular species, the formation of refractory organics, and the spectral changes induced in icy targets and in natural bitumens are described. In order to establish if the effects seen in the laboratory are in fact responsible for the surface properties of the TNOs, the present knowledge of the ion fluxes is reviewed. For objects at selected solar distances, dosage time is given versus depth into the material. As suggested by recent experiments, the contribution of the electronic energy loss and that due to knock-on collisions are given separately. The relevance of ion-irradiation for the physico-chemical properties of TNOs is demonstrated, and the need for future investigations is outlined. *To cite this article: G. Strazzulla et al., C. R. Physique 4 (2003).*
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Résumé

Irradiation ionique des OTNs : des flux mesurés dans l'espace aux expériences de laboratoire. On considère que l'irradiation ionique cosmique est un des processus qui gouvernent l'évolution des matériaux de surface des OTNs. Nous passons en revue la simulation en laboratoire des effets induits par l'irradiation dans des matériaux probablement constitutifs des OTNs. Nous décrivons en particulier la production de nouvelles espèces moléculaires, la formation de matériaux organiques réfractaires et les changements spectraux induits dans des cibles de glace et des bitumes naturels. Afin d'établir si les effets observés en laboratoire sont bien responsables des propriétés de surface des TNOs, nous rappelons l'état actuel de nos connaissances sur les flux ioniques. Nous donnons la relation entre temps d'irradiation et profondeur de pénétration pour des objets situés à diverses distances du soleil. Suivant une suggestion récente, la contribution des pertes d'énergie électronique et celle des collisions atomiques sont traitées séparément. Nous démontrons l'influence significative de l'irradiation ionique sur les propriétés physico-chimiques des OTNs et soulignons la nécessité de poursuivre ces recherches. *Pour citer cet article : G. Strazzulla et al., C. R. Physique 4 (2003).*
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1. Introduction

Knowledge of the physico-chemical properties of the surfaces of the small trans-Neptunian objects (TNOs) is scarce. Excellent reviews have been published on this subject. Among the most recent is that by Luu and Jewitt [1], which contains extensive references. Since the primary source of information on these interesting objects is their reflectance properties, there are a number of key questions that one would like to answer guided by laboratory data. These questions are all based on the fact that the surfaces of the TNOs are 'weathered' by impacts, by the solar UV, by energetic particle radiation and by thermal processing.

- Which materials and surface weathering mechanisms produce the observed spectral characteristics?
- Based on an understanding of the weathering process can predictions be made of new molecular species that formed in the surface layers?
- Is the chemical composition of the optical layers different from the internal layers from which most of the species emitted by comets come?
- What is the relationship of the TNOs with other objects in the Solar System such as Pluto, Triton, Centaurs, and short-period comets?

Based on solar system formation models, we have only a rough knowledge of the actual composition of the above objects and it is unclear whether they are similar. We also have only sparse information about the aging of the observed surfaces and the relative importance of the various weathering processes.

The complexity of the problem is such that analysis of the ground-based and space observations of the surfaces must be based on laboratory simulations that are used to model the effects of space weathering processes. Here we focus our attention on recent laboratory results on the effects induced by energetic ion irradiation of materials relevant to TNOs. To establish the extent to which laboratory results are relevant to clarify the open questions, we also discuss the recent observational evidence on the fluxes of cosmic ion populations bombarding the surfaces of those objects. Finally, in order to apply the laboratory results a description is given of the way the energy of incoming ions is released to the molecular species as a function of the depth into the target.

A number of studies have been done to investigate to what extent energetic processing (UV and/or energetic particle irradiation) can modify pre-existing grains, can modify grains that are reformed in the solar nebula and accrete to form larger bodies, can modify the whole object (e.g. [2–6]).

2. Laboratory experiments

Fast ions penetrating solids deposit energy in the target by elastic interactions with target nuclei and by inelastic collisions causing ionizations and excitations. Thus bonds are broken along the path of the incoming ion producing physico-chemical modifications and the erosion of material from the target (sputtering). The sputtering process and the related astrophysical applications will not be discussed here (for reviews see [2,3,7]). Chemical modifications induce the formation of molecules originally not present in the target. These molecules include species both more volatile and less volatile than the initially irradiated molecular mix. When carbon (or sulfur) is an important constituent of the irradiated material a refractory residue results which remains after warming the sample. That residue has a complex structure, which after prolonged irradiation evolves to form a hydrogenated amorphous carbon.

Among the techniques used by different groups to characterize the ion induced chemistry, particularly relevant are in situ IR (transmittance and reflectance) and Raman spectroscopy. IR spectroscopy is a powerful technique for studying chemical modifications induced by ion irradiation of frozen gases. It has been used by a number of groups to investigate effects induced in several ices and mixtures made of species bearing H, C, N, O and S atoms. Raman spectroscopy provides valuable information on the effects induced by fast ions impinging on solids, in particular on carbonaceous materials for which relevant information on the structural properties can be obtained [6].

2.1. Materials

The outer layers of the grains that initially formed the surfaces of the TNOs are thought to be covered by frozen volatiles. Therefore, a number of different ices and mixtures have been irradiated in different laboratories to study their chemical and/or structural evolution thermally and under irradiation. Usually frosts are made at low temperature (10–20 K) and their spectral characteristic recorded before, during and after ion bombardment. Targets are subsequently warmed-up and spectra taken versus increasing temperatures (20–300 K).

One problem is identifying the initial materials. Ideally we would know the composition of the surface of a given object at a given time of its evolution, such as at the time of its formation, and then follow the evolution induced by ion irradiation in order to establish if what we observe now on a TNO is like the material produced in the laboratory by the studied process. In reality we do not know the composition of the TNOs objects at the time of their formation nor do we know their present composition. Thus it is necessary to study targets whose composition mimics models of the initial composition or what we observe (with strong limitations) to be the present composition of TNOs.

The TNO materials must reflect, at least partially, the composition of pre-existing (interstellar) grains. Refractory dust particles (average radius $\cong 0.05$ μm) made of silicates or carbonaceous material are released from stars in whose atmospheres they are formed, into the interstellar medium. Occasionally diffuse clouds ($n_H \approx 1$–10^3 cm^{-3}, $T \approx 100$ K; where n_H is the total number of H atoms, i.e. H $+ 2$H$_2$, being H$_2$ the numeric density of hydrogen molecules) contract to form dense molecular clouds ($n_H \geqslant 10^4$ cm^{-3}, $T \approx 10$–20 K) where icy layers are formed on the grains. Dense molecular clouds, after further contraction are the places where stars and planetary objects are born.

Interstellar grains may have been 'reprocessed' in the solar nebula. Undoubtedly silicates (most probably olivines and pyroxenes), refractory carbons (amorphous carbons, polycyclic aromatic hydrocarbons, complex hydrocarbons, etc.), and ices are likely constituents of TNOs.

With the exception of water ice, no detection has been reported so far of other frozen molecular species on a TNO. Therefore, one irradiates those icy mixtures that are observed to be present on/in objects considered to be 'relatives' of TNO's such as short-period comets, Centaurs, Pluto and Triton. Short period comets may have originated in the Kuiper belt and formed beyond Neptune and Pluto. They seem to mimic rather well the composition of pre-stellar grains, i.e. ices are dominated by water, with significant contribution of CO$_2$, CO, methanol and other, even complex, hydrocarbons. The spectrum of the Centaur 5145 Pholus [8] has been matched with five components among which are 15% of water ice and 15% of methanol (or some other hydrocarbon). Pluto and the captured Triton could be the first-discovered TNOs. In addition to the dominant solid nitrogen (N$_2$) and solid methane, CO and CO$_2$ have been detected on Triton's surface [9]. Pluto has some CO, but less than Triton has, and no CO$_2$ [10].

Thus in addition to silicates and carbon, the icy materials that are of particular interest for TNO's objects are mixtures dominated by water and/or hydrocarbons and/or nitrogen. In particular nitrogen and water, because of their very different thermodynamic properties, are likely to be rapidly spatially segregated. In fact a careful analysis of the Pluto's spectrum [11] showed that the best model requires a surface mixture of three distinct units. The first is a thin, fine-grained, pure (i.e., segregated) CH$_4$ layer covering a compact polycrystalline substrate dominated by N$_2$ with small amount of CO (0.1–0.2%) and CH$_4$ (0.5%) that covers about 70% of the observed surface. The second unit (about 20% of the surface) is constituted by large-grained methane pure or covered by the N$_2$:CO:CH$_4$ mixture. The third unit (about 10%), bright and spectrally neutral, has been modeled with very fine grains of nearly pure nitrogen and is also compatible with the presence of tholins (a refractory organic material), suggested to explain the red slope in the visible spectrum. Solid carbon dioxide (CO$_2$) has not been detected. The detection of water (H$_2$O) is difficult because of the spectral contamination due to Charon whose surface is water ice rich. Recent near-IR spectra of Pluto, measured separately from Charon, have shown only very weak evidence of the existence of water ice [12]. The analysis of the near-IR spectrum of Triton leads to two possible representations of the surface [13]. The first is a surface with two distinct regions: a terrain made of dominant N$_2$ with traces of CO and CH$_4$, and a terrain made of a mixture of pure crystalline H$_2$O and CO$_2$ grains. The second possibility is that three regions are present, i.e., the same as above but with water and carbon dioxide grains spatially segregated.

With this in mind it is obvious that the number of ion irradiation experiments that need to be performed on relevant materials is enormous. Here we concentrate on those that concern the possible development of an organic crust on the surface of TNOs, the influence on the color of the surfaces, and the chemical composition. That is, one would like to predict the chemical species that should be present but have not yet been observed.

2.2. Results

2.2.1. The organic crust

Experimental results obtained in several laboratories demonstrate the production, starting from simple ices, of many new species including organic refractory materials [4–6,14]. In the solar nebula grains are, to some extent, 'reprocessed'. The irradiation history, during accretion, may be at least as important that in the ISM. This is particularly true if accretion occurred during an active phase of the young Sun (T-Tauri phase). Once formed, these objects were then exposed to the fluxes of solar, heliospheric, and galactic cosmic rays. Reviews of the effects expected on Oort cloud comets [15] and on Kuiper belt objects [16] have been presented. In particular it has been estimated that the external (0.1–0.5 m) layers of a comet were subjected to an irradiation dose of 600 eV/molecule. Deeper layers were subjected to a lower dose because the most abundant but less energetic ions are stopped by the external layers. Although the dose at depth is less than that suffered in the early evolutionary phases, it could be relevant to the behavior of comets. It has been suggested that a comet exposed to background particle
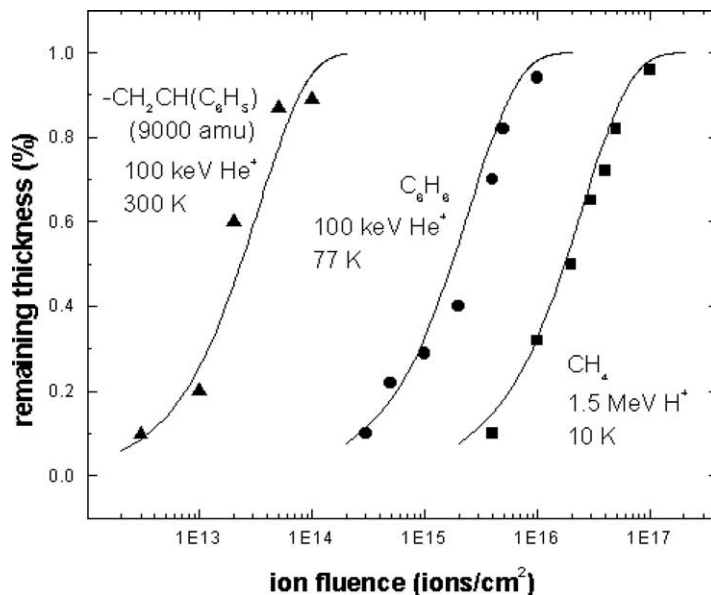
Fig. 1. The fraction of carbon incorporated into a refractory material is shown versus ion fluence for irradiated frozen methane (10 K), frozen benzene (77 K) and for polystyrene (9000 amu, 300 K). In the latter case the remaining fraction means the fraction of insoluble residue left over after irradiation. Data points have been fitted with an exponential function: $R = 1 - e^{\sigma\phi}$. Where ϕ (ions/cm^2) is the ion fluence and σ (cm^2) the cross section for the process. The cross sections of the process are 4×10^{-17} cm^2 for methane, 4×10^{-16} cm^2 for benzene and 3×10^{-14} cm^2 for polystyrene. These translate into G-values (see text) per carbon atom converted into refractory of 0.60, 1.3, 96 carbons per 100 eV deposited.

radiation obtains an outer web of non-volatile material which will lead to the formation of a substantial 'crust' [15]. The initial estimate of the thickness of the crust made by Strazzulla and Johnson [15] was based on the experimentally measured cross section ($\sim 4 \times 10^{-17}$ cm^2) for the conversion of frozen methane to a refractory residue [17,18]. Now the fraction of carbon incorporated into the refractory material has been measured for more cases. That fraction is given by an exponential function: $R = 1 - e^{\sigma\phi}$, where ϕ (ions/cm^2) is the ion fluence and σ (cm^2) the cross section for the process. In Fig. 1 the fraction of refractory carbon formed is shown versus ion fluence for irradiated frozen methane (10 K), frozen benzene (77 K) and for polystyrene (9000 amu, 300 K). In the latter case the remaining fraction means the fraction of insoluble residue left over after irradiation. The cross sections of the process are 4×10^{-17} cm^2 for methane, 4×10^{-16} cm^2 for benzene and 3×10^{-14} cm^2 for polystyrene. The results demonstrate that whatever the initial hydrocarbon is, it is converted to a refractory, insoluble residue. The cross section of the process depends on the molecular weight of the original material (it is about 10^3 times higher for polystyrene than for methane).

Due to irradiation processing, the TNOs could already contain organics with high molecular weight (e.g., heritage of pre-solar grains). In this case the crust formation process is much more efficient than in the case considered by Strazzulla and Johnson [15]. That is, the crust could be much thicker and/or have been developed much rapidly than previously estimated.

In situ Raman spectroscopy of carbon-containing ice mixtures reveal that the organic crust is already forming during bombardment at low temperature and does not require the warming of the sample [19,20]. This gives credit to the hypothesis that a comet crust can be already formed during a TNOs long residence far from the Sun and assures that crustal material is present on TNOs.

2.2.2. Spectral characteristics

TNO, Centaur, and other cometary nuclei exhibit very relevant color variations in the visual and near-infrared spectral regions from steep red to flat gray (e.g. [1]). These colors give a rough indication of the present composition of the surface which is determined by the alteration processes. One well-known model [21] has until recently explained the diversity of colors on different cometary objects as arising from competition between two principal processes: (1) cosmic ray irradiation assumed to produce reddening with increasing dosage; and (2) resurfacing by interplanetary dust impacts and collisions with other large bodies, which presumably excavate more pristine material with neutral colors from below the organic crust. Such a model is, in our opinion, too simplified. In fact some old laboratory ion-irradiation experiments demonstrated that fresh ices (e.g., methane) have flat spectra (and high albedo) that are reddened because of irradiation and, when further irradiated at higher doses these
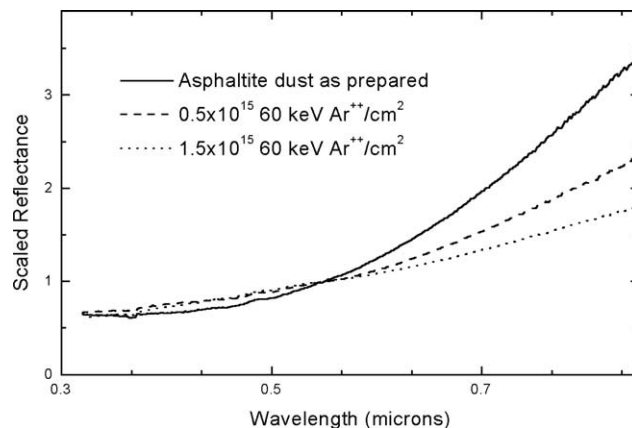
Fig. 2. Diffuse reflectance spectra (0.3–0.9 μm) normalized to 0.55 μm, from a sample of asphaltite (at 300 K) before and after ion irradiation. The results indicate that initially 'red' spectrum progressively flattens with increasing ion fluence.

spectra are flattened giving them a low albedo [22]. Those experiments have been done 'ex situ', i.e., by removing the samples from the scattering chamber and acquiring their spectra. This could alter, at least quantitatively, the results mainly because when extracted, the irradiated organics can react with atmospheric oxygen.

In the Catania (Italy) laboratory it has recently become possible to obtain 'in situ' diffuse reflectance spectra (0.6–2.5 μm) from samples that remain in the chamber throughout the irradiation processing and the subsequent reflectance measurements. The goal is to study relevant materials such as ice mixtures, silicates with and without icy and/or carbonaceous layers, and natural bitumens that have been suggested to be representative of the colors of TNOs [23].

In a first set of experiments an 'ageing' effect has been simulated by irradiating samples of natural red bitumens (asphaltite and kerite) with 30–60 keV ions. Their reflectance spectra were measured before and after irradiation. The results indicate that the initially 'red' spectrum characteristic of organics progressively flattens with increasing ion fluences as shown in Fig. 2. The laboratory spectra have been compared with astronomical spectra of TNOs. Moroz et al. [24] demonstrate that an observed variety of TNO spectral slopes can be reproduced by laboratory spectra corresponding to different ion fluences, i.e. to different exposure times in space.

2.2.3. Chemical composition

A large number of experiments done by different groups on several different icy species and mixtures indicate that ion irradiation, among other effects, induces the formation of molecules different from the irradiated ones (e.g. [2–7]). Thus some predictions can be made on which molecules might be expected to be present in the surfaces of TNOs although only a few have been observed at present. As stated above the icy mixtures that seem to be more relevant for TNOs are those observed in short period comets, believed to originate from the Kuiper belt region, of Centaur objects, and of Pluto and Triton believed to be objects belonging to the class of the TNOs.

One problem is to understand the different composition of comets and Pluto/Triton. Comets are water-rich with a composition, measured in the gas phase from sublimating molecules, that mimics quite well the mantles observed on icy grains in dense molecular clouds. In particular comets appear to be nitrogen poor. The surfaces of Pluto and Triton, on the other hand, are nitrogen rich although, locally, ice segregation could have formed surface spots rich in different molecules. Perhaps nitrogen diffuses from the internal layers of TNOs and is lost in space (sublimation) during the dynamical evolution from the Kuiper region to near by the Sun. Moreover the surface of at least the smaller among the TNOs and Centaurs could be composed primarily of a devolatilized skin (the crust).

Molecules, not yet observed, that can be predicted to be present on objects with surface areas rich in H_2O/CO_2 mixtures are carbonic acid (H_2CO_3) and carbon suboxides (C_3O_2 and C_3O) [25–28]. It has been also suggested that these species could be responsible for the CO distributed sources and for formaldehyde that has been detected in the laboratory mass spectrum of the sublimating residue as well as in cometary comae [25,27].

Experiments irradiating N_2 rich icy mixtures have been recently performed. Irradiation of mixtures $H_2O:N_2$ demonstrates formation of N_2O, NO and NO_2 [29]. Although nitrogen and water are likely to be rapidly spatially segregated, because of their very different thermodynamic properties, we believe that, due to seasonal sublimation and re-condensation on Pluto and Triton, the highly volatile nitrogen is deposited over the entire surface of the objects, i.e. including the water-rich spots. Experimental studies demonstrate that N_2 condensed on amorphous water ice diffuses in at temperatures of about 30 K and induces the

appearance of a water feature in the IR spectrum at about 1.88 μm that is easily distinguishable from the broad water band centered at about 2 μm [30].

Irradiation of binary mixtures $CO:N_2$ and $CH_4:N_2$ and of the ternary mixture $CO:CH_4:N_2$ demonstrates the formation of several new species among which carbon dioxide and suboxides, nitrogen oxides, and a number of CN bearing species [31]. These latter species are also produced for a large variety of mixtures containing H, C, O, and N bearing species and it has been suggested they could be responsible for the distributed CN emission in cometary comae [32].

With the above in mind it is likely that a plethora of not yet identified species have to be present because of ion irradiation of TNOs.

3. Measured fluxes of relevant ion populations

The energetic ion environment in the outer solar system is composed of components from multiple sources. In the work of Cooper et al. [33,34] the time-averaged spectra of H, He, and CNO (mostly O) ions have been compiled from three years (1997 to 2000) of solar energetic particle (SEP) measurements by the Advanced Composition Explorer (ACE) in L1 halo orbit near Earth, from measurements for the same ion species by the Voyager 1 and 2 spacecraft in the outer solar system during the mid-1994 epoch of solar minimum, and from a one-dimensional model for inward transport of anomalous cosmic ray (ACR) ions from the solar wind termination shock and galactic cosmic ray (GCR) ions from the Local Interstellar Medium (LISM). The Ultra-Low Energy Isotope Spectrometer (ULEIS), Solar Isotope Spectrometer (SIS), and Cosmic Ray Isotope Spectrometer (CRIS) instruments on ACE are the primary data sources near 1 AU, while Voyager 1 and 2 fluxes are taken from the Cosmic Ray (CRS) experiments on each spacecraft. For protons the SEP, ACR, GCR, and summed contributions are shown in Fig. 3 from the Cooper et al. [33] version of this model for 40 AU, the inner boundary of the classical Kuiper Belt.

The SEP spectra for each ion group (H, He, CNO) are determined at ACE's position from power-law fits to ULEIS and SIS data. In fits for long-term averages of SEP fluxes the power-law indices are very similar for different ions [35]. Most of these fluxes accumulate from large solar flares and coronal mass ejection events in the 1997–2000 rise from minimum to maximum solar activity. SEP ion flux is not directly measured in the present flux model beyond 1 AU but is scaled as the inverse square of solar distance from the ACE spectra. At 40 AU the SEP H^+ ions dominate the total flux only below 1 MeV. SEP fluxes may actually be lower than here estimated at this distance and beyond, due to energy losses associated with propagation through the magnetic field in the outwardly expanding solar wind.

The ACR fluxes essentially vanish during epochs of maximum solar activity, again due to energy loss in the solar wind, and have their highest intensity in the Ecliptic during every other 11-year solar cycle, e.g., the last ACR maximum being for the 1987 solar activity minimum. During low solar activity in the mid-1990s the ACR ion intensities measured at Voyager [36–38]
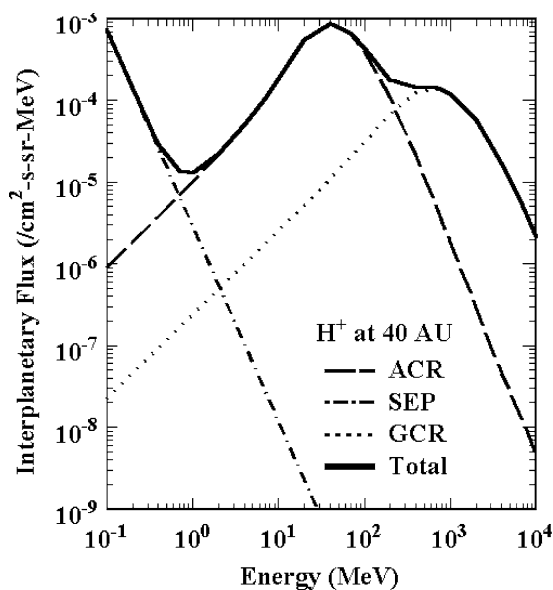

Fig. 3. Total and component proton flux spectra at 40 AU from the model of Cooper et al. [33,34] for solar energetic particles (SEP), anomalous cosmic ray (ACR) ions, and galactic cosmic ray (GCR) ions.
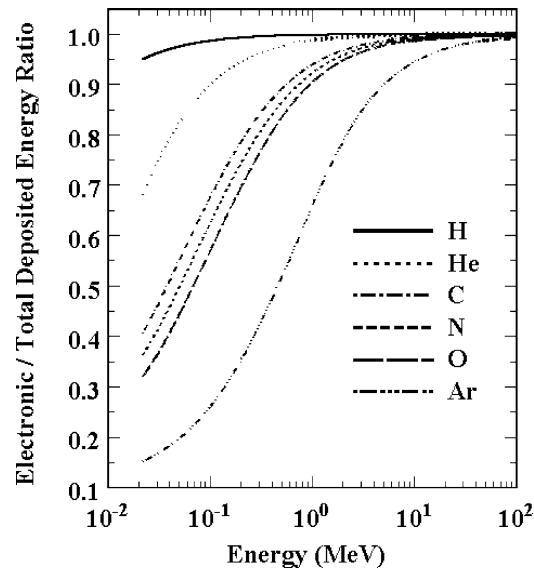
Fig. 4. Ratio of electronic to total energy loss as a function incident energy for various ions incident on H_2O water ice target. Computed from the Stopping and Range of Ions in Matter (SRIM) code of Ziegler et al. [40] at http://www.SRIM.org/.

were at intermediate levels, so we presently use this epoch for approximation of intensities averaged over multiple consecutive solar cycles. The ACR fluxes at 40 AU and elsewhere are computed using the cosmic ray transport model with parameters determined from 1-AU fits to SIS and CRIS data from ACE, CRS data from Voyager, and model spectra for source fluxes of the ACR ions at the 85-AU termination shock during minimal solar activity in mid-1994 as given by the Voyager CRS publications cited just above.

The GCR ion flux spectra are determined for transport model parameter fits for peak (solar minimum) fluxes to CRIS data from ACE and to CRS data from Voyager. The H^+ fluxes in Fig. 4 roll off downward at energies below a few hundred MeV/n, due to energy loss in the solar wind, while the power-law ($\sim E^{-2.75}$) declines with increasing energy E at GeV/n energies. This arises from the relatively loss-less transport of LISM ions into the heliosphere at such energies. The ion source spectra in the LISM are assumed to fit a $\sim p^{-2.75}$ power-law [39] at all energies above 0.1 MeV/n in terms of ion momentum p, consistent with origins from acceleration by supernova shocks in interstellar space within the Galaxy.

A significant limitation of the present work is that the Cooper et al. [33] flux model did not include plasma and suprathermal ions at energies less than 0.1 MeV/n. Higher energy ions deposit energy primarily by electronic ionization (Fig. 4), but elastic nuclear collision losses increasingly become important at lower energies. Cooper et al. [33] do include low energy protons in their flux models but the model data have not yet been released for publication. This means that dosage rates quoted in the present work at small penetration depths, less than one μm, are lower limits to what would be calculated by extending energy coverage downward with flux model data from the later Cooper et al. work.

4. Energy deposition versus depth and chemical change

The exposure of the surfaces of TNOs to energetic particle radiation leads to chemical and physical alterations as discussed. These are produced down to the depth of penetration of the most energetic ions or electrons. This depth is determined by the energy lost per unit path length to the solid, dE/dx. Whereas the character of the changes produced does not change significantly for the various types of incident particles, the efficiency of production and the depth of the effect strongly depends on the particle type and energy. The amount of change is often given as a function of dose, the energy deposited per unit volume in the material by the incident radiation as described above. Particular chemical changes are, in turn, often characterized by G-values, the amount of a product produced per 100 eV energy deposited by the incident particles. For example, the amount (yield) of frozen organic converted to chain carbon by radiolysis can be expressed as a G-value. G-values are primarily appropriate when back reactions are not important as in the example mentioned. That is, when there is a vacuum interface, as there is on the surface of a TNO, H_2 once formed at $T \gtrsim 20$ K escapes so back reactions can not occur. When the relevant material is confined so that even volatile species cannot escape (for instance $CO_2 \rightarrow CO + O$ at $Y \lesssim 30$ K), then G-values are only meaningful at low doses. Further, G-values are often treated as 'constants' that are measured for one incident species and assumed to apply for

all. However, they can depend on the energy per unit volume deposited and not simply on the total energy deposited as often assumed. Therefore, the radial spread of the deposited energy about the ions' track and its penetration depth affect the amount of chemical change produced by an individual ion. Therefore, in spite of the fact that radiolysis has a long history, it is important to understand how energetic particles deposit their energy in solids in order to use and extrapolate the available data sets for application to particular solar planetary or astrophysical problems.

In Fig. 1 the ability to form a crust was represented by a cross section, not a G-value. Multiplying the x-axis by the dE/dx of the incident ion gives the total amount of energy deposited per unit volume near the surface, D, the dose. The fits in Fig. 1 would now be written $[1 - e^{-D/D_0}]$ where D_0 is the characteristic dose for producing the chemical change: $D_0 = [dE/dx]/\sigma$. In a similar manner the damage cross sections can be converted to a G-value: $G^{-1} = D_0 V$, where V is the volume of the relevant target species. The dE/dx is often given as an energy loss per unit mass, in which case V is replaced by the relevant species mass. G-values, as well as cross sections, are given for crust formation in the caption to Fig. 1.

A very energetic ion loses energy to the grains in the regolith of a TNO by ionizations and excitations of the atoms and molecules, as stated above. The rate of this loss with depth is given by the electronic stopping power, $(dE/dx)_e$. For a very fast ion, this energy loss can be small on entrance into the material, but at the end of the ion's path the deposition rate is rapid and there is a region in which the energy density deposited is always quite high, the Bragg peak. In this region both the electronic and knock-on (elastic) loss contribute so the net effect can be significant although in a small volume. Low energy solar wind ions ($\sim$1 keV/amu) lose their energy both in electronic excitations and ionizations and in knock-on collisions. They do not penetrate significantly, as indicated earlier, and the energy densities deposited are relatively high locally, like in the Bragg peak at the end of the path of the fast particles. Therefore, as stated above, in many respects the nature of the damage and chemistry induced is similar for very different incident ions although the distribution with depth and efficiency can differ significantly. In Fig. 4 the ratio of electronic to total energy loss as a function incident energy for various ions incident on H_2O water ice target is reported. Those ratios have been computed from the Stopping and Range of Ions in Matter (SRIM) code [40].

Space weathering of TNOs by energetic particles occurs over long time scales which can be quantified from available models for average fluxes of these components onto the TNO surfaces in different regions of the heliosphere and the LISM. The model flux data for 40 AU in Fig. 5 provide estimates for irradiation environments of TNOs in the classical Kuiper Belt at 40–50 AU. There is also a scattered disk population of TNOs on highly eccentric orbits with aphelia distributed out in the far outer heliosphere, e.g., to the 85 AU distance of our model termination shock and even further out to 1000 AU in the Very Local Interstellar Medium (VLISM) environment approximated by the LISM spectra in Fig. 5.

In Fig. 6 we show, as a function of depth to one cm, the times in years needed to accumulate total ($H + He + CNO$) electronic and elastic nuclear dosages $\sim$100 eV/16-amu sufficient for significant radiolytic change in the irradiated material. The dosages
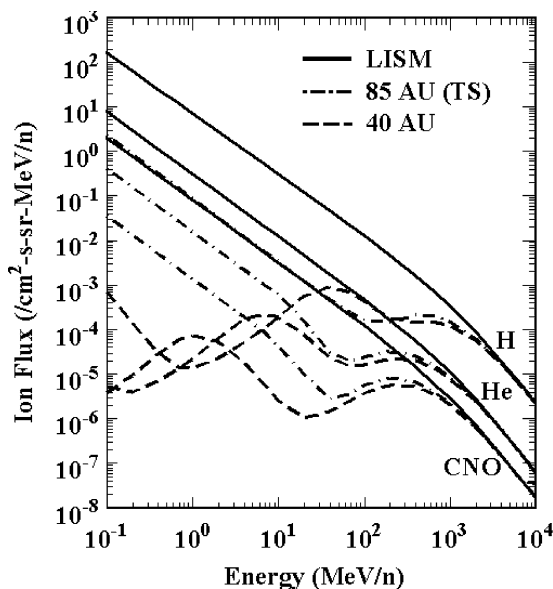

Fig. 5. Model ion fluxes as function of energy per nucleon (MeV/n) for cosmic ray H, He, and CNO (mostly O) ions at 40 AU, an assumed solar wind termination shock at 85 AU, and in the Local Interstellar Medium. These flux spectra are taken from Cooper et al. [33], except that the Anomalous Cosmic Ray (ACR) components of spectra at the termination shock in that work were plotted incorrectly and are given here for heliospheric conditions near solar minimum in mid-1994 from the Voyager analyses [36,37].
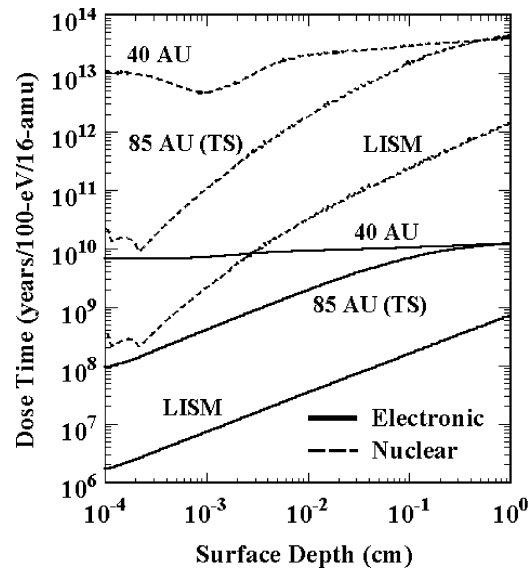
Fig. 6. Times in years at 40 AU, 85 AU (termination shock), and in the LISM to accumulate chemically significant dosages of 100 eV per 16-amu as functions of target depth for H_2O ice at unit density. The solid and dashed curves are respectively for the electronic ionization and elastic nuclear collision components (Fig. 4) of ion energy loss. Total dosages from all ions are computed by the methods of Cooper et al. [41] from summation of dosages for the individual H, He, and CNO (mostly O) spectra in Fig. 3. Ion incident angles are 47 degree to approximate isotropic omnidirectional incidence onto a flat surface.

are computed by the method of Cooper et al. [41] only for energy losses of the primary incident ions and not for any secondary nucleons, X-rays, or gamma rays that might arise from inelastic nuclear collisions or from electromagnetic interactions. These secondary radiation components tend to deposit energy at water ice depths greater than several cm at unit density, so effects of the primary ions are best seen in the first cm. This is also nearly the maximum depth that composition can be measured by remote spectroscopic imaging for all TNOs known to date. Decreasing radiolytic dosage time (and increasing dosage rate) at small depths arises from increasing differential flux at lower energies in Fig. 5. The dosage profile at 40 AU is relatively flat with depth, since the model ion spectra at this distance are dominated by higher energy, more penetrating ions. The low energy portions of the model flux spectra rise (and the dose times significantly decline) at 85 AU, due to increasing contributions to local fluxes from ACR ion acceleration at the termination shock. In the LISM the shortest dosage times are computed, since interstellar shock acceleration produces relatively large fluxes of ions outside the heliosphere and far beyond the modulating effect of the solar wind. Thus the minimum dosage time at μm depths for significant radiolytic effects with $G \sim 1$ (e.g., Fig. 1) is 10^{10} years at 40 AU and 10^8 years near the termination shock. An upper limit on dosage time of 2×10^9 years was given by Cooper et al. [33] for secondary nuclear and electromagnetic interactions of high energy galactic cosmic rays, as computed from the CREME96 model [42] for GCR (but not SEP or ACR) flux spectra and total irradiation dosages near 1 AU. The relatively flat dosage profiles for 40 AU in Fig. 6 indicate that additional irradiation from SEP or ACR ions is not significant for this distance at the inner edge of the classical Kuiper Belt, but the ACR ions clearly increase in importance further out, and dosage rates would increase with eccentricity and aphelion distance for scattered TNOs.

5. Remarks and future studies

Dose scales for the occurrence of the effects discussed in Section 2.2 have to be converted to time scales using the results shown in Fig. 6. This allows one to evaluate if a particular effect is relevant on a given astronomical object and to what depths it is relevant.

The chemical effects (Section 2.2.3) begin to occur on a dose scale of the order of a few eV/16 amu [2–6,25–32] and are roughly dependent on the total (electronic plus elastic) energy deposited by ions into the target. Thus, starting from the (unknown) initial composition of the icy species on a TNO object, noteworthy quantities of newly synthesized species are formed on relatively short time scales as a consequence of its interaction with cosmic ray ions (see Fig. 6). As an example: to accumulate a total dose of 10 eV/16 amu at the surface of a TNO, times lower than 10^9, 10^7, 10^5 yrs for objects at 40 AU, 85 AU, or in the LISM respectively are necessary. At that dose a water-carbon dioxide mixture will have suffered chemical

alteration and CO/CO_2 and H_2CO_3/CO_2 ratios of 0.2 and 0.1 respectively are expected [25]. Scattered objects are irradiated less uniformly with depth than the classical objects (see Fig. 6). Thus they are expected to have a composition not uniform with depth (0–1 cm) into respect to the classical objects.

A different question is the possible observation of molecular species that are presently very difficult to detect, since even components that are predicted to be dominant, such as water ice are not readily observed. Effects, such as the formation of an organic crust or color changes further complicate the observation of newly formed species as they mask to some extent the underlying layers. Newly synthesized molecules may have a better chance to be observed in objects that might initially have been TNOs, such as Pluto and Triton, the Centaurs, or as gaseous species in cometary comae.

The development of an organic crust due to the conversion of low molecular weight hydrocarbons into a web of bound molecular species occurs on a dosage scale of the order of 100 eV/16 amu [15]. The total amount of deposited energy can again be used to estimate the time scale relevant to TNOs. From Fig. 6 it is clear that on time scales of the order of the age of the solar system an outer web of devolatilized material is developed down to substantial depths, especially for comets in the Oort clouds, but also for scattered TNOs and marginally for classical Kuiper's objects.

The question of the color is more complex and requires the acquisition of many more experimental results. The materials, the textures, and the processes that determine the variety of observed colors are presently difficult to take fully into account. Different formation scenarios are possible. If we assume that simple hydrocarbons converted into an organic crust are mainly responsible for the observed color, then it is necessary to go into the lab and study the color variation as a function of irradiation dose and initial composition. As stated above this is part of an ongoing program at Catania. If the hypothesis that already complex materials are accreted on TNOs (e.g., bitumen like), the initial experiments [43] seem to indicate that color changes are produced more efficiently by ions with a significant elastic stopping power. This demonstrates the importance of separately estimating the deposition, by cosmic ions, of elastic and inelastic energy as in Fig. 6. It has been in fact observed that a very red bitumen (asphaltite) is darkened at dosages of the order 100 eV/16 amu mainly because of elastic interactions. To evaluate the relevance of the process from Fig. 6 is not yet possible because plasma and suprathermal ions at energies less than 0.1 MeV/n have not been included. This means that dosage rates at small penetration depths, from which most of the 'information' on colors come from, are only lower limits.

We want to stress once again the importance of separately evaluating the contribution of energy deposition via elastic interactions. A number of the chemical or physical changes recently studied, as, e.g., the discussed color change of asphaltite, seem to be produced much more efficiently by the elastic energy deposited, which would be a new and interesting result. Some of those effects are presently studied in the laboratory and are potentially interesting for astrophysical objects such as TNOs. These effects include the formation of hydrogen peroxide from irradiated water ice [44,45] and the amorphization of crystalline silicates [46–48].

Acknowledgements

References

[1] J.X. Luu, D.C. Jewitt, Annual Review Astronomy and Astrophysics 40 (2002) 63–101.
[2] R.A. Baragiola, R. Vidal, M. Shi, W. Svendsen, J. Schou, D. Bahr, Nucl. Instr. Meth. Phys. Res. (2003), in press.
[3] R.E. Johnson, R.W. Carlson, J.F. Cooper, C. Paranicas, M.H. Moore, M. Wong, in: F. Bagenal (Ed.), Jupiter: Planet, Satellites, and Magnetosphere, Cambridge University Press, 2003, in press.
[4] M.H. Moore, R.L. Hudson, P.A. Gerakines, Spectrochimica Acta A 57 (2001) 843–858.
[5] G. Strazzulla, in: Y.J. Pendleton, A.G.G.M. Tielens (Eds.), From Stardust to Planetesimals, 1997, p. 423.
[6] G. Strazzulla, G.A. Baratta, M.E. Palumbo, Spectrochimica Acta A 57 (2001) 825–842.
[7] R.E. Johnson, in: B. Schmitt, C. de Bergh (Eds.), Solar System Ices, Kluwer Netherlands, Dordrecht, 1998, p. 303.
[8] D.P. Cruikshank, T.L. Roush, et al., Icarus 135 (1998) 389–407.
[9] D.P. Cruikshank, T.L. Roush, T.C. Owen, T.R. Geballe, C. de Berg, B. Schmitt, R.H. Brown, M.J. Bartholomew, Science 261 (1993) 742–745.
[10] T.C. Owen, T.L. Roush, D.P. Cruikshank, J.L. Elliot, L.A. Young, C. de Berg, B. Schmitt, T.R. Geballe, R.H. Brown, M.J. Bartholomew, Science 261 (1993) 745–749.
[11] S. Doute', B. Schmitt, E. Quirico, T.C. Owen, D.P. Cruikshank, C. de Berg, T.R. Geballe, T.L. Roush, Icarus 142 (1999) 421–444.
[12] W.M. Grundy, M.W. Buie, Icarus 157 (2002) 128–138.

[13] E. Quirico, S. Doute, B. Schmitt, C. de Berg, D.P. Cruikshank, T.C. Owen, T.R. Geballe, T.L. Roush, Icarus 139 (1999) 159–178.

[14] G. Strazzulla, in: B. Schmitt, C. de Bergh (Eds.), Solar System Ices, Kluwer Netherlands, Dordrecht, 1998, p. 281.

[15] G. Strazzulla, R.E. Johnson, in: R. Newburn Jr., M. Neugebauer, J. Rahe (Eds.), Comets in the Post-Halley Era, Kluwer Netherlands, Dordrecht, 1991, p. 243.

[16] R. Gil-Hutton, Pl. Sp. Sci. 50 (2002) 57–62.

[17] G. Foti, L. Calcagno, K.L. Sheng, G. Strazzulla, Nature 310 (1984) 126–128.

[18] G. Strazzulla, L. Calcagno, G. Foti, Astron. Astrophys. 140 (1984) 441–444.

[19] G. Strazzulla, G.A. Baratta, R.E. Johnson, B. Donn, Icarus 91 (1991) 101–104.

[20] G. Strazzulla, G.A. Baratta, Astron. Astrophys. 266 (1992) 434–438.

[21] J. Luu, D. Jewitt, Astron. J. 112 (1996) 2310–2318.

[22] G. Andronico, G.A. Baratta, F. Spinella, G. Strazzulla, Astron. Astrophys. 184 (1987) 333–336.

[23] L.V. Moroz, G. Arnold, A.V. Korochantsev, R. Wäsch, Icarus 134 (1998) 253–268.

[24] L.V. Moroz, G. Baratta, E. Distefano, G. Strazzulla, L.V. Starukhina, E. Dotto, M.A. Barucci, Earth Moon Planets (2003), in press;
L.V. Moroz, G. Baratta, G. Strazzulla, L.V. Starukhina, E. Dotto, M.A. Barucci, E. Distefano, Icarus (2003), submitted.

[25] J.R. Brucato, M.E. Palumbo, G. Strazzulla, Icarus 125 (1997) 135–144.

[26] J.R. Brucato, A.C. Castorina, M.E. Palumbo, G. Strazzulla, Pl. Sp. Sci. 45 (1997) 835–840.

[27] M.H. Moore, R.K. Khanna, Spectr. Acta 47 (1991) 255–262.

[28] P.A. Gerakines, M.H. Moore, Icarus 154 (2001) 372–380.

[29] L. Ottaviano, M.E. Palumbo, G. Strazzulla, in: S. Aiello, A. Blanco (Eds.), What are the Prospect for Cosmic Physics in Italy?, 2000, p. 149.

[30] M.E. Palumbo, G. Strazzulla, Can. J. Phys. 81 (2003) 217–224.

[31] M.H. Moore, R.L. Hudson, Icarus 161 (2003) 486–500.

[32] G. Strazzulla, Sp. Sci. Rev. 90 (1999) 269–274.

[33] J.F. Cooper, E.R. Christian, R.E. Johnson, in: Proc. 27th Intern. Cosmic Ray Conf. Hamburg, Aug. 7–15, Vol. 10, 2001, p. 4267.

[34] J.F. Cooper, E.R. Christian, J.D. Richardson, C. Wang, Earth Moon Planets (2003), in press.

[35] R.A. Mewaldt, G.M. Mason, G. Gloeckler, E.R. Christian, C.M.S. Cohen, A.C. Cummings, V. Davis, J.R. Dwyer, R.E. Gold, S.M. Krimigis, R.A. Leske, J.E. Mazur, E.C. Stone, T.T. von Rosenvinge, M.E. Widenbeck, T.H. Zurbuchen, in: R.F. Wimmer-Schweingruber (Ed.), Solar and Galactic Composition, in: AIP Conf. Proc., Vol. 598, AIP, Melville, NY, 2001, p. 165.

[36] A.C. Cummings, E.C. Stone, Sp. Sci. Rev. 78 (1996) 117–128.

[37] A.C. Cummings, E.C. Stone, Sp. Sci. Rev. 83 (1996) 51–62.

[38] E.C. Stone, A.C. Cummings, W.R. Webber, J. Geophys. Res. 101 (1996) 11017–11026.

[39] W.R. Webber, M.S. Potgieter, Astrophys. J. 344 (1989) 779–785.

[40] J.F. Ziegler, J.P. Biersack, U. Littmark, The Stopping and Range of Ions in Solids, Pergamon Press, New York, 1985.

[41] J.F. Cooper, R.E. Johnson, B.H. Mauk, H.B. Garrett, N. Gehrels, Icarus 149 (2001) 133–159.

[42] A.J. Tylka, et al., CREME96: A revision of the cosmic ray effects on micro-electronics code, IEEE Trans. Nucl. Sci. 44 (1997) 2150–2160.

[43] L.V. Moroz, L. Starukhina, G. Strazzulla, G. Baratta, E. Dotto, M.A. Barucci, G. Arnold, Icarus (2003), submitted.

[44] G. Strazzulla, G. Leto, O. Gomis, M.A. Satorre, Icarus 164 (2003) 163–169.

[45] O. Gomis, M.A. Satorre, G. Strazzulla, G. Leto, Pl. Sp. Sci. (2003), in press.

[46] K. Demyk, Ph. Carrez, H. Leroux, P. Cordier, A.P. Jones, J. Borg, E. Quirico, P.I. Raynal, L. d'Hendecourt, Astron. Astrophys. 368 (2001) L38–L41.

[47] C. Jäger, D. Fabian, F. Schrempel, J. Dorschner, Th. Henning, W. Wesch, Astron. Astrophys. 401 (2003) 57–65.

[48] J.R. Brucato, G. Strazzulla, G.A. Baratta, Astron. Astrophys. (2003), in press.