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CHARGED PARTICLE TRACKS IN POLYMERS: Number 2 – Registration of Heavy Ions During the Flight of Gemini VI

by

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ABSTRACT

Several small disks of cellulose nitrate plastic were included as a part of a dosimetry package aboard the Gemini VI spacecraft for the purpose of recording the heavy component of the primary cosmic radiation. An average of 10 tracks/cm²-day was found in the plastic detectors. Most of these were judged to be tracks of heavy recoil ions; about 1 track/cm²-day was judged to be due to the primary cosmic ray particles.

SUMMARY

The Problem

Conventional dosimeters do not accurately measure the dose from the multicharged component of the primary cosmic radiation. Since the contribution of such radiation to the total dose received will be considerable during the long range manned space flights, it is important to develop a heavy ion dosimeter for this purpose.

The Findings

An average of 10 tracks/cm²-day was found in the plastic detectors. Most of these were judged to be tracks of heavy recoil ions; about 1 track/cm²-day was judged to be due to the primary cosmic ray particles. Polymer detectors are best suited for registration of slow, heavy ions. For polar orbits or flights outside the vicinity of the earth, separation of heavy ion flux into (dE/dx) intervals and a subsequent estimate of the dose can be made.

Recommendations

Further development of polymer detectors is recommended. Improvement of surface characteristics and sensitivity, as well as longer duration equatorial and polar flights are clearly desirable.

INTRODUCTION

Cosmic rays above the earth's atmosphere produce about 25 mrad dose/24 hours or about 10 rads/year. The proton and alpha particles constitute all but about 1 per cent of the total particle flux. Nevertheless the remaining one per cent of heavy particles are responsible for about 40 per cent of the total dose. Most of the dose is produced through the action of slow, heavy $(Z \ge 6)$ nuclei.

Probably the main feature of these heavy nuclei is their high (dE/dx) (rate of energy loss). The heaviest particles can produce up to a maximum of 100,000 ion pairs per micron of tissue, or a dose in excess of 1000 rads to a single cell of 10 microns diameter.⁽¹⁾ This is about 50 times the maximum value produced by a proton. The mode of action of such densely ionizing radiation has never been investigated to the point where its biological effectiveness relative to proton or electron beams could be established. Since direct damage to cellular systems have been observed for single heavy particles (effect on hair follicles, etc.), it becomes important to investigate the long term action of this type of radiation which is clearly different from our natural radiation environment. Thus, any manned space flight in excess of several weeks duration should contain heavy particle dosimeters

capable of defining the radiation environment within the vehicle. This cannot be accomplished with conventional dosimeters designed for monitoring the electron, proton and X-ray component of the cosmic ray environment.

Within the past two years a number of dielectric solids have been found which can serve as heavy charged particle detectors.^(2,3) Tracks are delineated in these materials as a result of a preferential removal, by etching, of the high chemical reactivity material along the particle trajectory. The most sensitive materials found to date are polymers. Cellulose nitrate plastic is one of the most sensitive of these, requiring for track formation a rate of energy loss, (dE/dx), by the incident particle to be above $2 \frac{MeV}{mg/cm^2}$. Tracks of alpha particles with (dE/dx) greater than $2 \frac{MeV}{mg/cm^2}$ are observed as well as other particles having the (dE/dx) in excess of this minimum value. For particles with relativistic velocities only those with $Z \ge 26$ have a (dE/dx) above this value.^(2,3) Several other materials with different sensitivities are known to exist,⁽³⁾ and these together with cellulose nitrate can be used as low resolution (dE/dx) spectrometers. Subsequent estimates of the dose due to heavy cosmic ray particles can be made.

Polymer detectors have a number of characteristics that make them particularly attractive for space applications. They are light, rugged, compact, insensitive to temperature and humidity changes, have no latent image fading at ordinary temperatures and are simple to process. Since protons are not registered, the very long exposures needed to

record adequate numbers of heavy ions can be performed. Polymer detectors have the advantage of low weight over electronic systems, e.g., scintillation telescopes. A complete package can be made to weigh a fraction of one ounce. The system avoids a problem encountered with sensitive nuclear emulsions which during prolonged exposures become completely saturated with proton tracks, making it impossible to distinguish the heavy ion tracks from the background. The severe problems of latent image fading encountered with the use of the less sensitive emulsions during prolonged exposures is also avoided.

The orbit characteristics of the Gemini and the early Apollo flights are such that the geomagnetic cutoff for charged particles varies from about 5 to 10 BeV/amu. Thus, only particles with relativistic velocities reach the space craft.

EXPERIMENT

Packets consisting of 10 mil cellulose nitrate plastic and a 600 micron thick Ilford G.5 nuclear emulsion were assembled as in Fig. 1. Both the plastics and the emulsion were in the form of 16 mm diameter disks wrapped and sealed in heavy black paper. Five packets were distributed inside the Gemini VI capsule while the sixth was used as a ground control. The shielding varied from 1 gm/cm^2 for the packets were mounted on the hatch to several gm/cm^2 for the others. The packets were



IO MIL GELLOLOSE NITRATE SHEET

Fig. 1 Edge-on view of the polymer detector packet.

exposed to incident radiation during the 17 orbit flight of Gemini VI lasting 25.9 hours in an orbit with 28.9 degree inclination and having average perigee and apogee of 100 and 162 miles, respectively.

DISCUSSION

Track Registration

In Fig. 2 is shown the relation between the (dE/dx) and the atomic number Z of particles in cellulose nitrate with $\beta(=v/c)$ of 0.145 and 0.899, corresponding to particles with energies of 10 MeV/amu and 1200 MeV/amu respectively. The data were obtained from a computer rangeenergy program for heavy ions in materials of known composition.^{*} The $\beta = 0.145$ curve corresponds to the low energy part of the cosmic ray spectrum present at high latitudes, but absent in the regions traversed during the flight of Gemini VI. The $\beta = 0.899$ curve represents the (dE/dx) of particles with relativistic velocities and is valid for higher β since the (dE/dx) of a particle does not change by more than 10 - 14 per cent of its value at $\beta = 0.899$. The horizontal line, (dE/dx)_{min.} = 1.2 $\frac{MeV}{mg/cm^2}$, represents our experimental determination of the minimum value of (dE/dx) for track registration in cellulose nitrate. Figure 2 suggests that the lightest particle observable in

* To be published.





$$\left(\frac{\text{MeV}}{\text{mg/cm}^2}\right)$$
 as a function of the atomic number Z of particles

with $\beta = (v/c) = 0.145$ and 0.899 in cellulose nitrate.

cellulose nitrate for $\beta = 0.145$ is carbon and for $\beta = 0.899$ is either manganese or iron.

In order for a track to be observable through the technique of chemical etching the particle must have a (dE/dx) which is above some minimum value that is characteristic of the material. (2,3) However, it is felt by the authors that this is a necessary but may not be a sufficient condition for track registration; energy distribution must also be considered. The energy deposition per unit volume along the trajectory is a function of the particle velocity. Particles of relatively low velocity lose a major fraction of their energy through many, low energy transfer collisions, thus confining this energy to a very small volume along the trajectory. This is contrasted with the case of relativistic particles that lose energy through fewer, high energy transfer electron collisons; the struck electrons, having considerable range, distribute the energy over a much larger volume. Thus, the energy per unit volume deposited along the particle trajectory is significantly less for a relativistic than for a low velocity particle, even though both may have the same (dE/dx). As an example, one can consider an alpha particle with $\beta = 0.036$ and an iron nucleus with $\beta = 0.899$. Although both have the required (dE/dx) for track registration in cellulose nitrate, i.e., 1.25 MeV the former areas mg/cm readily observable while the latter are not expected to register in its cellulose nitrate. For track registration of relativistic particles in cellulose nitrate a considerably greater Z than 26 may be required; a)

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The concept of $(dE/dx)_{min.}$ for a given material as presented by Fleisher, et al.,⁽²⁾ appears to be useful for low velocity heavy ions, but may need modification when applied to relativistic particles.

The observed track length, \mathcal{L} , of a particle track in a polymer is a function of the etch rates along the particle trajectory, r_{T} , and the bulk etch rate of the polymer, r_{B} . For a particle incident at an angle Θ to the surface \mathcal{L} is given by

$$\mathcal{K} = \int (\mathbf{r}_{\mathrm{T}} - \mathbf{r}_{\mathrm{B}} \csc \Theta) \mathrm{dt}$$

where t is the time of etch. The above also holds for a particle traversing the entire thickness of the detector, e.g., a relativistic particle. If $r_{\rm R}/(r_{\rm B}\csc\Theta) > 1$, then ℓ should increase with etch time. Therefore, it is desirable to have the ratio $r_{\rm T}/r_{\rm B}$ as large as possible. It was found that $r_{\rm T}/r_{\rm B} \cong 20$ for fission fragment tracks in cellulose nitrate using 30 per cent NaOH etch solution at 60° C. At 23° C, $r_{\rm T}/r_{\rm B}$ increases to over one hundred. Thus fission fragment tracks are easily observable, with the measured range closely corresponding to the theoretical range of the particles. For particles with (dE/dx) approaching the (dE/dx)_{min} of the given material it appears that $r_{\rm T}/r_{\rm B}$ becomes small, approaching unity, and tracks become more difficult to delineate.

A method of increasing r_T/r_B in cellulose nitrate has been found. In Fig. 3 is shown the observed track length, ℓ , as a function of the



Fig. 3 Observed track length, λ , as a function of temperature of etchant (30% NaOH) for 2.1 MeV alpha particles in cellulose nitrate. The theoretical range is 10 microns.

etchant temperature for 2.1 MeV alpha particles. It is seen that by etching at low temperatures for longer etch times (~100 hours) ℓ is increased and at 0°C approaches the theoretical range of the particles. This effect is also demonstrated in Fig. 4a,b. Here are shown tracks of ⁴⁰Ar ions in cellulose nitrate. The particles having energies of 10.4 MeV/nucleon enter the picture at the right. The tracks are about 70 microns long and represent only the initial portion of the total range, which is 181 microns. Tracks in Fig. 4a,b were etched for 13 min at 60° C and 57 hr at 2°C, respectively. It is noted that tracks etched at lower temperatures have a narrower cone angle suggesting that $(r_{\rm T}/r_{\rm B})_{2}\circ_{\rm C} > (r_{\rm T}/r_{\rm B})_{60}\circ_{\rm C}$. The ratio $r_{\rm T}/r_{\rm B}$ can be even further increased by stimulating the diffusion of etchant in the long, narrow cavity of the track by the use of ultrasonic vibrations (see Fig. 5).

Results

Upon recovery, the plastics and emulsions were given a standard processing and examined with the aid of an optical microscope. In plastics the tracks were observed at regular etch intervals. The number and positions of heavy ion tracks were noted in each. In emulsion, track width and delta-ray density were used as track identification criteria.

The mean geomagnetic cutoff for the orbit was estimated to be 6.3 $GeV/amu.^{(4)}$ Assuming the mean thickness of material surrounding the detector was 10 gm/cm² of aluminum, the expected incident heavy ion



Fig. 4a,b Initial 70 micron portions of 10.4 MeV/nucleon ⁴⁰ Ar ion tracks in cellulose nitrate. Particles traveled from right to left. Tracks in (a) and (b) were etched for 13 min at 60°C, and 57 hrs at 2°C respectively, in a 30% NaOH solution. Tracks in (a) have a cone angle of about twice that of tracks in (b), suggesting a different rT/rB for the two etch processes. The sharp terminal end of the track indicates that further etching would increase the track length. Note that the surface of the polymer is pitted in (a), making track observation difficult.



Fig. 5 Track of 10.4 MeV/nucleon ⁴⁰ Ar ion in cellulose nitrate. The particle entered from the right. The track was obtained by etching for 2 hrs at room temperature in a 30% NaOH solution immersed in an ultra-sonic bath. The track is 145 microns long; the theoretical range is 181 microns.

flux on the detector for particles with $Z \ge 20$ was estimated to be about 4.1 ions/cm²-day.⁽⁵⁾ Examination of emulsions yielded 3.0 tracks/cm²-day, a value within the limits of experimental error. An average of 10 tracks/cm²-day was found in the plastic detectors. The track length varied from 5 to 35 microns. Most of the tracks were short, about 10 - 15 microns, and were judged to be due to either imperfections in the polymer surface which upon etching gave track-like appearance, or heavy recoil ions resulting from the interaction of incident protons and the nuclei of the emulsion-plastic interface. About 1 track/cm²-day was judged to be due to the primary cosmic ray particles. These were the longest tracks found and were judged to be too long for recoils. Upon repeated etching they maintained sharp endings in the polymer while increasing in length with etch time. These tracks may possibly be the result of heavy particles, such as the C.N.O group, that have been highly degraded in energy by the material of the space craft, or they may be due to relativistic particles with Z greater than 26. Because of the very short exposure time and limited detector area (10 cm²) only a few tracks were available. Several of them were steeply dipping tracks, thus further analysis was not possible.

CONCLUSIONS

Polymer detectors are best suitable for registration of slow, heavy $(Z \ge 2)$ ions (see Figs. 4a,b and 5). With a possible exception of very heavy particles ($Z \ge 26$), most relativistic particles do not appear to possess sufficient (dE/dx) for track formation in cellulose nitrate.

A number of changes and improvements in the techniques have suggested themselves as a result of this flight:

1. Longer exposures are clearly necessary. For near equatorial orbits, where the heavy ion flux $(Z = \geq 20)$ is approximately 4 ions/cm²-day, exposure periods of several weeks are desirable. The two to three week duration Gemini missions would probably suffice. Even longer exposures could be obtained by leaving the detectors aboard the Agena target vehicle and subsequently recovering them during the next mission.

2. Improvement in the surface characteristics of the detectors is desirable. Some imperfections in the surface are etched preferentially and sometimes have track-like appearances. Long etch times, necessary for delineation of long tracks, tend to degrade the polymer surface, making track observation and photomicrography difficult.

3. A study is being made to determine what Z a relativistic particle must have in order to produce a track in cellulose nitrate.

4. Examination of Fig. 2 reveals that the (dE/dx) of a relativistic iron nucleus is some 30 times greater than that of a comparable

velocity proton. Since nuclear emulsions record "minimum ionization particles." there exists at present a range of about three decades between the most sensitive emulsion and the most sensitive polymer detector. In fact the sensitivity of cellulose nitrate roughly corresponds to the sensitivity of Ilford K minus 1 nuclear emulsion. which is one of the least sensitive emulsions manufactured. (6,7) It seems very improbable that the sensitivity of polymer detectors could be increased to the point where registration of minimum ionization tracks would be possible. However, a sensitivity increase by a factor of 2 or 3 does not appear improbable and would be highly desirable. In fact the $(dE/dx)_{min.}$ of 1.2 $\frac{MeV}{mg/cm^2}$ for cellulose nitrate quoted by us differs from the value of $2 \frac{MeV}{mg/cm^2}$ given earlier by Fleischer, et al.⁽²⁾ The $(dE/dx)_{min}$ of a material appears to be a function not only of the material itself but also of the etching process. The type of etchant and the temperature both play a role in determining the etch rate along the particle trajectory and the bulk etch rate and thus determine the $(dE/dx)_{min}$ of the material. With an optimum combination of polymer material and etching process, it is probable that a detector of greater sensitivity can be achieved.

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