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Chemical Bonds Broken in Latent Tracks of Light Ions in Plastic Track Detectors

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Abstract

When a swift ion is slowed down through a plastic detector it creates a latent track. In nuclear track detectors, this latent track can be specifically etched by an appropriate chemical solution. This enlargement process is due to a higher etch velocity (V_T) along the ion's path than in the non-damaged part of the detector. The etched track velocity is definitely linked to the damage created by the incoming ion in the detector material. A relationship between the physical parameters of the energy deposition and the variation in this etched track velocity with the ion energy cannot easily be explained. We present here our study on the chemical damage created by several ions in a cellulose nitrate type detector and our first attempt to simulate them by the use of the hit theory.

Etched Track Velocity and Physical Parameters

In previous studies attempts were made to outline the kind of relationship that exists between the measured etched track rate, V_T , and the basic physical parameters of the ion–matter interaction. In cellulose nitrate (LR115, Kodak) as well as poly allyl diglycol carbonate (CR-39, Tastrak) irradiated with MeV alpha or lithium ions, the average radial delta ray dose, the total LET or REL_{350} do not uniquely describe the experimental V_T s [1–3]. (Figure 1)

Chemical Damage and Infra Red Spectroscopy

The LR115 used in this study is a trademark of Kodak; the detector is 12 μm thick. Cellulose nitrate (CN) constitutes 90% of its weight, the residual 10% are additives. The degree of CN nitration used is around 2.3. Each irradiated sample consists of a stack of several LR115 detectors each 12 μm thick (from 17 to 32 detectors according to the ion energy). The energy loss in each layer was estimated using the TRIM code [5]. After exposure to known ion fluences in the vacuum, the detectors were analyzed by infrared (FTIR) spectroscopy

Figure 2 presents the cellulose nitrate structure as well as the main modifications observed by infrared spectroscopy [7]. The band at 1160 cm^{-1} can most likely be attributed to the glycosidic bond (C–O–C) that links two pyranose rings [8]. The band at 1600 cm^{-1} corresponds to the O–NO₂ bonds, and the one at 1340 cm^{-1} to the C–H bonds.

Chemical Bond Breaking Cross Sections

Here, it is assumed that the relative decrease in the considered bond (Π_{bond}) can be described by [9]:

$$\Pi_{\text{bond}} = \exp^{-\sigma F}$$

where F is the fluence of the incoming ions and σ is defined as the global scission cross section corresponding to the final effect observed after chemical equilibrium.

Figures 3 and 4 show the measured scission cross sections plotted against the initial ion energy for boron and carbon ions that were slowed down in the LR115.

In each case the same tendency can be observed: the cross sections decrease from C–O bonds to N–O and to C–H bonds. The scission cross sections increase inversely in relation to the ion energy.

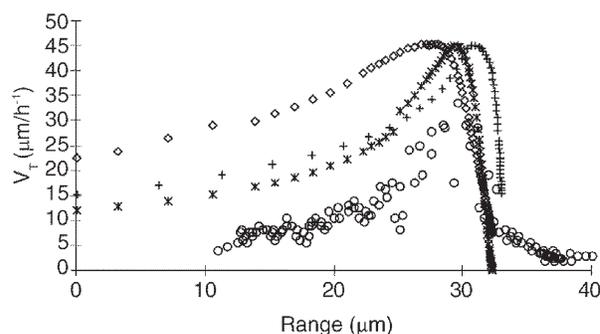


Figure 1. Comparison of the etched track rate (V_T) for a Li ion in (○) CR-39 [3] based on experimental data with, (*) the average dose from δ rays within 8 nm of the ion's path [4], (◇) total LET [5] and (+) REL_{350} [6].

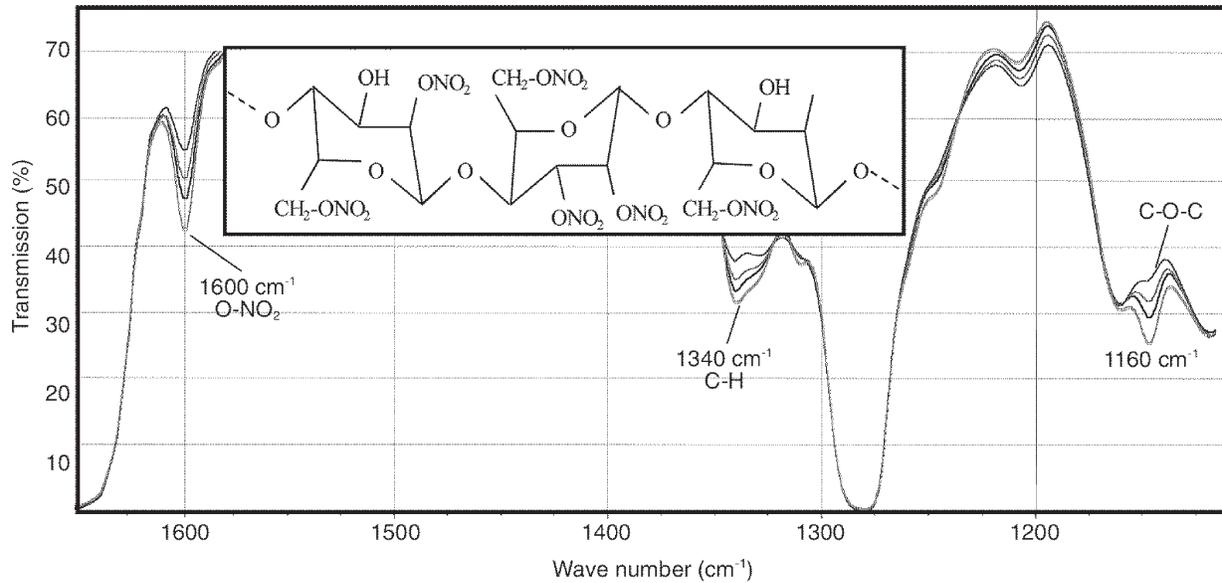


Figure 2. Main modifications (band decreasing) observed by FTIR (transmission mode) during irradiation of a 12 µm thick LR115 with ^{241}Am .

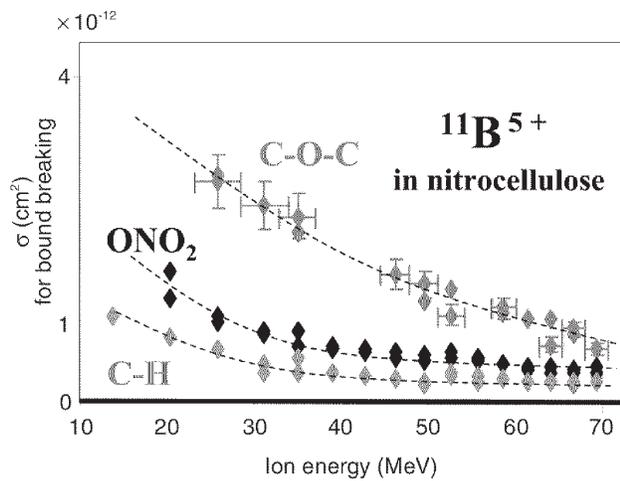


Figure 3. Scission cross sections for $^{11}\text{B}^{5+}$ in LR115.

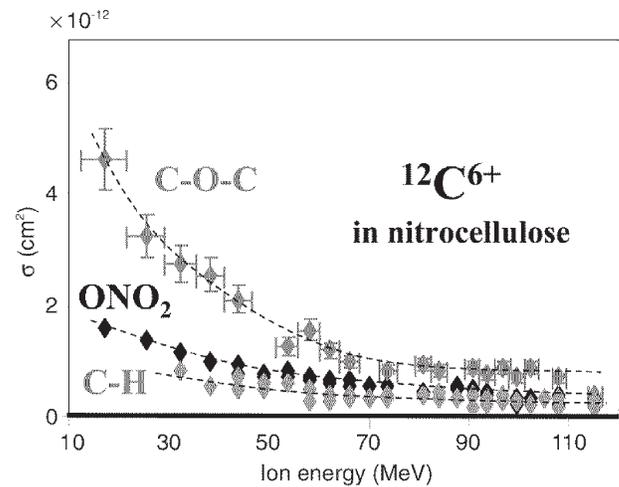


Figure 4. Scission cross sections for $^{12}\text{C}^{6+}$ in LR115

Table 1. Simulated and experimental scission cross sections.

Ion	Energy (MeV)	Experimental σ_{CO} (cm ²)	Model σ_{CO} (cm ²) $m = 1$ $a_0 = 0.2 \text{ nm}$ $D_0 = 2 \times 10^5 \text{ Gy}$	Experimental σ_{NO} (cm ²)	Model σ_{NO} (cm ²) $m = 1$ $a_0 = 0.2 \text{ nm}$ $D_0 = 3 \times 10^5 \text{ Gy}$
$^{11}\text{B}^{5+}$	69	6.7×10^{-13}	5.5×10^{-13}	3.9×10^{-13}	3.7×10^{-13}
	49	1.5×10^{-12}	7.0×10^{-13}	5.9×10^{-13}	4.6×10^{-13}
	26	2.7×10^{-12}	9.8×10^{-13}	1.0×10^{-13}	7.0×10^{-13}
$^{12}\text{C}^{6+}$	102	6.7×10^{-13}	9.3×10^{-13}	3.0×10^{-13}	5.8×10^{-13}
	80	9.5×10^{-13}	1.0×10^{-12}	4.0×10^{-13}	7.4×10^{-13}
	17	4.6×10^{-12}	2.2×10^{-12}	1.6×10^{-12}	1.5×10^{-12}

Dose and Chemical Structure

To try to describe the chemical structure of the latent track, the same approach [10] as proposed by Katz and Pinkerton [11] is developed for the nuclear emulsion response. Thus it is stipulated that the probability P of breaking a given chemical bond is:

$$P(r) = \{1 - \exp[-\bar{D}(r)/D_0]\}^m$$

where m is the number of hits needed to break a bond, D_0 the dose at which there is one hit per target and $\bar{D}(r)$ the average local dose delivered to the bonds at a distance r from the ion's path (deposited by free electrons emitted by the incident ion). Such a dose quantity concept is proposed by Miterev [12] in literature describing the relationship between V_T and the ion parameters. The dose $\bar{D}(r)$ is calculated using the following simple expression averaged over the target size a_0 [13]:

$$D(r) = \frac{N Z^2 e^4}{m c^2 \beta^2} \left(\frac{1}{r^2} - \frac{1}{r \cdot \tau} \right)$$

This relation gives the dose deposited by the different generations of electrons produced by the incident ion as a function of the distance r from the ion's path, where N is the number of target electrons per unit volume, e the elementary charge, m the electron mass, c the speed of light, τ the range of the most energetic electron from an ion moving at relative speed β , and Z the effective charge of the ion [14]. More sophisticated solutions for calculating this dose have been proposed more recently [15]. At this stage, however, precision is not sought, but a definition is wanted of the principles of our approach.

Finally, the cross section for bond breaking is expressed by:

$$\sigma = 2\pi \int_0^\tau P(r) r dr$$

where a_0 and D_0 and m are parameters fitted using the experimental data. The same set of D_0 , a_0 , and m must, however, describe the chemical damages observed for all the different ions. Assuming that one hit (one electron) can break a bond, m is taken as equal to 1. Figure 5 outlines this model.

Table 1 gives some results obtained from the loss of the nitrate functions and the C-O bond breaking.

Conclusion

Table 1 shows the first attempt to describe, with the present model, the experimental chemical cross sections with a target size of 0.2 nm and D_0 values of 2×10^5 Gy and 3×10^5 Gy for C-O and N-O bonds respectively. At this stage any differences in the initial electron energy spectra have been neglected. In spite of the use of the very simple calculation of dose $D(r)$, it is surprising to

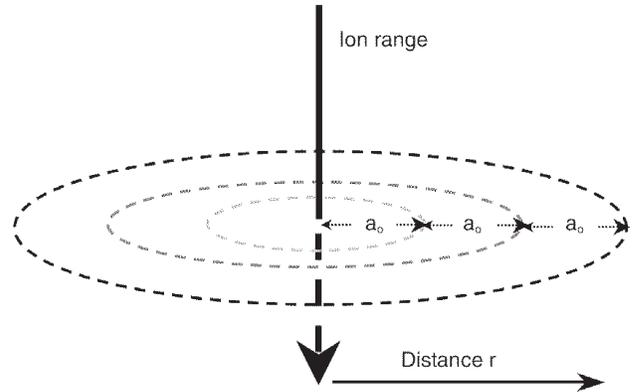


Figure 5. Our model assumptions in calculating the average delta ray dose in ranges of radial distances a_0 .

$$\sigma = 2\pi \left\{ \int_0^{a_0} P(r) r dr + \int_{a_0}^{2a_0} P(r) r dr + \dots + \int_{na_0}^{\tau} P(r) r dr \right\}$$

find a reasonable agreement between the experimental and calculated values. However, the model also shows a significantly weaker dependence on ion energy as compared to the experimental observations (see also Figure 1).

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