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Studies on the nuclear tracks in CR-39 plastics

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Abstract

A review was given for our recent studies on the latent tracks in CR-39 nuclear track detector. The radial size of track core has been determined through UV spectral measurements combined to the model of track overlapping and by AFM observations of slightly etched detectors. The track core radius was found to be about a few nano-meters and almost proportional to the cubic root of stopping power. As a control study, the etching properties of CR-39, irradiated by low-LET radiation, has been examined. The observed depth-dependence and dose-rate dependence of the bulk etch rate of the irradiated CR-39 were explained that the damage formation process was governed by the reaction between the radiation induced radicals and the oxygen supplied from the air. This indicated that latent tracks in CR-39 are produced through local radiation induced oxidation process along the ion paths. Studies on vibration spectra, Near-IR, FT-IR and Raman spectra, of CR-39 have been also carried out to estimate the chemical structure of the latent tracks. The creation of OH group in irradiated CR-39 has been confirmed.

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1. General introduction

The latent track formation in CR-39 plastics as a result of the slowing down of an ion and many secondary electrons ejected around the path will be discussed. From the viewpoint of the track physics, the radial dose distribution around the ion trajectory is an essential study (Waligorski et al., 1986). However, it is difficult to discuss the properties of the latent track in terms of the dose distribution only. The energetic electrons lead not only to new chain ends but also other chemically reactive sites in the polymer (Fleischer et al., 1975). Some chemical process should follow the physical one.

Through a study on the track registration in an out-gassed CR-39 under vacuum, it was found that the reaction between the radiation induced radical and the dissolved oxygen play an important role in the formation of the permanent damage (Fujii et al., 1987). This primitive work has revealed not only the decrease of etch pit size by the storage under vacuum before the irradiation, but also the partial recovery of the size by a rapid air-leak after the irradiation subsequent to the long-term storage under vacuum. The published papers implied the following model for track formation (Henke et al., 1970; Drach et al., 1987a, b; Fujii et al., 1987; Csige et al., 1991). A highly ionizing particle breaks polymer bonds, leaving free radicals that either react with dissolved oxygen in CR-39 to form a permanent latent track or recombine, failing to form a latent track, depending on the concentration of oxygen near the ion's path. Increasing the oxygen concentration increases the fraction of radicals that react. Using this model we have attained at least two components of radicals with life-time of about 40 minutes and a few minutes (Yamauchi et al., 1992). The considered model explained the observed results well, but we knew little about the radicals, the latent track, the oxygen dissolved and other things that appears in this model. In other word, we understood little about the track chemistry in CR-39.

In this decade my colleagues and I have been engaged in the study on the latent tracks in CR-39 detector, as well as calibration studies for light ions (Yamauchi et al., 1995a, b, 1996, 1997, 1999b, 2001b). Our studies on the latent tracks were divided in three parts. The first part is the studies on size of track core radius (Yamauchi et al., 1999c, 2000, 2003a, c). An atomic force microscope (AFM) and UV-visible spectrometer were applied in this series of studies. The radii of track core were in about a few nano-meters and almost proportional to the cubic root of the stopping power. In

the second part, the bulk etching properties of CR-39 irradiated by low-LET radiation has been examined to determine the dose-response relation (Oda et al., 1997; Yamauchi et al., 1999a, 2001a, d, 2003b). The results have indicated that the damage in CR-39 was produced through the local radiation induced oxidation. In the third parts, vibration spectra, Near-IR, FT-IR and Raman spectra, of the irradiated CR-39 have been obtained to elucidate the chemical structure of the latent tracks (Oda et al., 1997; Yamauchi et al., 1999c, 2000, 2001c, 2002, 2003c). The creation of OH group has been confirmed in the irradiated CR-39, whose concentration would be related to the rate of the penetration of etching agent along the latent track.

2. Track core size

2.1 Introduction

Conductometry has been an effective and commonly applied method to evaluate the radial track etch rate in various kinds of thin films (Apel et al., 1998, 1999; Bean et al., 1970; Peterson and Enge, 1995). Because the radial track etch rate reflects the radial dose distribution, the method could be useful not only to evaluate the track core size as the first approximation but also to examine the damage distribution around ion path (Enge, 1995). On the other hand, FT-IR spectral studies, combined to the dose-model, have been made on ion irradiated cellulose nitrate to measure bond scission cross sections (Barillon et al., 1999; 2000). Because these methods require thin films as specimens, it is difficult to apply them to CR-39, which is very brittle. Different methods for the size estimation of latent track were reviewed well in the literature (Enge, 1995). Unfortunately, we could not find a suitable method for CR-39 there.

It has been revealed that UV-visible spectra of the irradiated CR-39 were sensitively dependent on the absorbed dose and ion fluence, as well as ion type and energy (Oda et al., 1997; Yamauchi et al., 1999c, 2000). The fluence dependence of the UV spectra was successfully interpreted by a model of latent track overlapping. This means that the track core size in CR-39 can be evaluated from a series of UV spectra based on samples exposed to different particle fluences. This is the first method (called the UV method) in the present study. It was found to be applicable even for light ions like proton and He.

As a new imaging tool, atomic force microscopy (AFM) has been

demonstrated to have a much higher spatial resolution than that of traditional optical microscope (Drndic et al., 1994; Yamamoto et al., 1999; Yasuda et al., 1998, 1999). Recently, Yasuda has proposed a new method to assess the radial size of track cores in CR-39 using AFM (Yasuda et al., 2000, 2001). In the method, the growth curves of pit radius in nano-meter dimension were obtained, by observing the mouth size of etched tracks after extremely short-time etchings. Extrapolating the growth curves toward the beginning of etching, the coordinate at the intersection was regarded as the track core size. Although this method is similar to the method using SEM (Bohus et al., 1989; Enge, 1995), it has a great advantage that the surface observations can be made in air. Yasuda and the present author are making a collaborative study to improve the AFM method (Yamauchi et al., 2003a).

2.2 Model of track overlap

In this model it is assumed that each latent track has identical chemical structure and acts as an equivalent color center. At ranges of fluence where track overlap is negligible, the absorbance of the irradiated CR-39 will be proportional to the fluence, because the area occupied by tracks is also proportional to it. On the other hand, if the overlapping is significant, the area occupied by tracks is not proportional to fluence, moreover, the chemical structure and the optical property in the overlapped region will be different from that of a single track. As a result, the proportional relation between the absorbance and the fluence will break. Therefore, it is possible to determine a fluence where the proportional relation is broken. In the followings we call this fluence as the critical fluence. The method to determine the critical fluence from the UV-visible spectra has been given (Yamauchi et al., 1999c, 2000, 2003a).

Let us make a consideration about the track accumulation process in a unit area of 1cm^2 . So the number of tracks, n , is equal to the fluence. The real track has a complex structure along the path and also in radial direction. The track is, however, expressed by a cylinder whose length is equal to the ion range in the present model. In the top view, the track is a simple circle with an area s . On this assumption, the track accumulation process is almost the same as the wetting process of the ground by drops of rain. The occupied area or fraction by tracks at a fluence of n is written as $A(n)$. The overlapped region should be subtracted relevantly from the simple summation of track

area of ns . We shall introduce a simple approximation as follows: The occupied area at a fluence of $n-1$ is $A(n-1)$, and then the residual pristine area becomes $1-A(n-1)$. If the next ion was dropped in the occupied area, the area is assumed not to increase. And if it was outside the area, the occupation increases by s . The probability of the former case is equal to $A(n-1)$, and then that of the later is $1-A(n-1)$. Therefore, the expected occupied area at a fluence of n can be written as:

$$A(n) = A(n-1) + s\{1 - A(n-1)\}. \quad (1)$$

Solving this recurring formula with an initial condition of $A(1) = s$, we obtain

$$A(n) = 1 - (1-s)^n. \quad (2)$$

Because the track section, s , is sufficiently small to be compared with the considered unit area, this can be expressed as:

$$A(n) = 1 - \exp(-sn). \quad (3)$$

In Fig. 1, the calculated track occupation fraction are shown using Eq. (3). For example, in the case of a track radius of 1 nm, the surface is almost fully occupied by tracks at a fluence of 2×10^{14} ions/cm². As expected from basic consideration, the surface is occupied at lower fluence as increasing the track core radius. The validity of these calculations was demonstrated well by simulations (Yamauchi et al., 2000).

The occupied area consists of different regions with various folding of tracks. Eq. (3) can be expanded as,

$$A(n) = \{sn\} \exp(-sn) + \left\{ \frac{(sn)^2}{2} \right\} \exp(-sn) + \left\{ \frac{(sn)^3}{6} \right\} \exp(-sn) + \cdots. \quad (4)$$

The first term in the right is the area in tracks without overlapping, the second term is double overlap or 2 folds region and the third is 3 folds region. The area of N fold region is expressed as:

$$A_N(n) = \left\{ \frac{(sn)^N}{N!} \right\} \exp(-sn). \quad (5)$$

It is not difficult to derive this equation directly. In Fig. 2, the occupied area by tracks $A(n)$, the area of single $A_1(n)$, the double $A_2(n)$, the triple $A_3(n)$, the ten folds $A_{10}(n)$ and the 50 folds $A_{50}(n)$ are shown against the fluence at a track radius of 1 nm.

Because the track overlapping is a statistical phenomenon, it is not possible to find a fluence where the early overlap happens. The overlapping can happen even at a fluence of 2 ions/cm² by chances. So we should express the critical fluence as a certain

band on the fluence and also the center value as representative. We defined that the band starts at the fluence where double overlap becomes significant, and ends where the triple becomes so. And the center value in the band was selected as the critical fluence. In this manner, the relation between the critical fluence and the typical track core radius was obtained, as shown in Fig. 3. Complete spectral data related to the critical fluences were given in the literature (Yamauchi et al., 1999c, 2000).

2.3 Track radius evaluated from the UV and AFM methods.

The obtained track core radii by the UV method have been summarized as a function of the stopping power for the light ions as followings:

$$r_t = 0.519 \left(\frac{dE}{dx} \right)^{0.39}, \quad \text{in nm}, \quad (6)$$

where dE/dx is in the unit of keV/ μm (Yamauchi et al., 2000, 2003a). Some studies have also insisted that the core radius in some materials was proportional to the cubic root of the stopping power (Albrecht et al., 1985; Tombrello, 1984). Further examination for various ions must be made to know the general relation between the core radius and the stopping parameters.

Recently we have found that the results from the UV method is in agreement with that from the AFM method within experimental error (Yamauchi et al., 2003a). The examined stopping power was up to 10000 keV/ μm . This means that the curves by Eq. (3) can also be used in higher region of the stopping power.

3. Effect of low-LET radiation

3.1 Introduction

Many studies have been made on the bulk etch rate of CR-39 irradiated by high doses of gamma-rays (Akber et al., 1980; Green et al., 1982; O'Sullivan et al., 1982; Zamani et al., 1985; 1986; Portwood and Henshaw, 1986; Charvat and Spurny, 1988; Sharma et al., 1991; Shweikani et al., 1993; Shinha et al., 1997; Fazal-ur-Rheman et al., 2001). In these works the examined ranges of the absorbed dose were controlled to be comparable to the local energy deposition inside ion tracks.

3.2 Dose, dose-rate and depth dependence

We have pointed out that the bulk etch rate increased exponentially with the absorbed dose under a certain dose-rate and surrounding conditions (Oda et al., 1997). Even with a same total absorbed dose, the irradiation at higher dose-rates resulted in lower bulk etch rates. The exponential relation was observed also in the vacuum, but the bulk etch rate was much less sensitive to the dose than that in air. These observations suggested strong influence of the oxygen in forming the damage, therefore we have investigated the dependence of the bulk etch rate on the dose-rate under various conditions (Yamauchi et al., 1999). At dose-rates above 50 Gy/s, the dose dependence of the bulk etch rate was found to be almost equivalent to that in the vacuum. We have proposed a model, based on the experimental results, in which the damage production rate was assumed to be proportional to both the dose-rate and the concentration of oxygen. It was also hypothesized that the oxygen was consumed during the damage production and the feed rate of oxygen from the surroundings was proportional to the difference between the oxygen concentration in CR-39 under equilibrium condition and that under irradiation. The observed dose-rate dependence was explained well by this model where the decrease in the bulk etch rate at higher dose-rates should be caused by the reduction of oxygen level in CR-39 during irradiation.

The bulk etch rate was usually determined from both the etching duration and the radius of etched track of high-LET particles like fission fragments which indicates the thickness of layer removed. This is an effective method as far as the etched track is cylindrical in side view and the bulk etch rate is constant. Recently, we have found the “Bottle-like structure” in the etched tracks of fission fragments in the gamma-irradiated CR-39 (Yamauchi et al., 2001a). The etched track had double stages in radius, larger near the surface and smaller in the deeper region along the common axis of the trajectory, as shown in Fig. 4. The two parts were connected with a thin transition layer, so that the etched track looked like an upper part of a wine bottle. The parts near the surface with larger radius corresponded to the “body” of the bottle and the other part in the deep with smaller radius was equivalent to the “bottle neck”. Such a unique etched track was possible when the bulk etch rate increased only in the surface region where the “body” was formed. The above mentioned dependence of the bulk etch rate on the absorbed dose, the dose-rate and the surrounding conditions have been determined for this surface region. On the other hand, the “neck” was formed in the deeper layer. The

etching properties of this region were found to be the same as that for CR-39 irradiated in vacuum, fully out-gassed. This implied that the concentration of dissolved oxygen in the deeper layer reached zero during gamma-irradiation. Therefore the damaged region near the surface was inferred to relate the amount of the oxygen fed from the surrounding air. Then we have obtained the dose-rate dependence of the thickness of the damaged region in detail (Yamauchi et al., 2001d). The thickness was found to decrease with increasing the dose-rate. At higher dose-rates, the oxygen supplied on the surface should be soon consumed in forming the damage and it will become more difficult for the oxygen to diffuse toward deeper layers.

The property of bulk etching of CR-39 irradiated by low-LET radiation was expressed by experimental formulas. The samples were Japanese CR-39 of BARYOTRAK, and the etching was made in 6N KOH solution at 70 °C. The bulk etch rate in the damaged region, V_{irra} , at absorbed dose, D in Gy, and constant dose rate, R in Gy/s, were expressed as follows:

$$\begin{aligned}\frac{V_{irra}}{V_b} &= \exp(gD), \\ g &= g_{ox} + g_{vac}, \\ g_{ox} &= \frac{4.8 \times 10^{-5}}{R + 0.2}, \quad g_{vac} = 9.0 \times 10^{-6},\end{aligned}\tag{7}$$

where V_b is the bulk etch rate of unirradiated CR-39, g is a constant in Gy^{-1} . The factor g consists in two components; the first is related with oxygen amount, g_{ox} , and the second is independent of oxygen amount, g_{vac} (Yamauchi et al., 1999a). On the other hand, the bulk etch rate in vacuum, V_{vac} , and in deeper layer, where “neck” was formed, was given as:

$$\frac{V_{vac}}{V_b} = \exp(g_{vac}D).\tag{8}$$

As shown in this equation, V_{vac} is independent of the dose-rate. The thickness of the damaged region, d^* , is given by:

$$d^* = 5.03 R^{-0.353}, \quad \text{in } \mu\text{m}.\tag{9}$$

The thickness is not dependent on the total dose. The present author has suggested the possibility of applying CR-39 to high-dose gamma-ray dosimeter, by evaluating both the bulk etch rate and the thickness of the damaged region (Yamauchi et al., 2001d).

3.3 Model for radiation-induced oxidative degradation

In order to explain the bulk etching properties of the CR-39, a model has been proposed in which the damage was assumed to be formed through the reaction between radiation induced free radicals and the oxygen dissolved (Yamauchi et al., 2002b). At first, the production rate of the radical, or the active site in the polymer, was hypothesized to be proportional to the dose rate, R . Secondly, the radical has some probability to vanish naturally, which might relate recombinations. This assumption is based on the track registration study on the well out-gassed CR-39 (Yamauchi et al., 1992). Finally, the rate of the damage formation was postulated to be proportional to the concentration of the radical, $[P\bullet]$, and the oxygen concentration, C . Therefore, the following mass balance equation was derived for the concentration of the radical:

$$\frac{d[P\bullet]}{dt} = \Phi R - k_1[P\bullet] - k_2[P\bullet]C, \quad (10)$$

where Φ is the specific rate constant for the radical formation, k_1 and k_2 are the rate constant for radical decay and oxidation or fixation of the radical, respectively. Under a steady condition, it becomes as follows:

$$[P\bullet] = \frac{\Phi R}{k_1 + k_2 C}. \quad (11)$$

Since the damage density is proportional to $[P\bullet]C$ and the irradiation duration, t , a uniform damage distribution in depth would be expected in the region where the oxygen exists, provided that k_1 is much less than $k_2 C$. The observation on the bottle-like etched track indicated that the damage distribution should be uniform in the damaged region near the surface. So it is possible to re-construct the damage distribution using this model. If the cross-linking is dominant, the second term on the right hand side in Eq. (10) is replaced by the square of $[P\bullet]$ multiplied by a constant, and such a uniform distribution could not be expected.

On the other hand, the following equation was derived from the mass balance of the dissolved oxygen,

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - k_2[P\bullet]C, \quad (12)$$

where D symbolizes the diffusion coefficient of oxygen in CR-39 and x is the depth

measured from the center of the film.

As the boundary conditions on the oxygen concentration at the surface of CR-39, two different cases were examined. The first was the constant condition in which the concentration at the surface is constant during the irradiation. The second was the surface resistance condition, in which the rate of transfer of diffusing oxygen through unit area of the surface is proportional to the difference between the actual concentration in the surface, C_s , at any time and the equilibrium concentration, C_o . The equilibrium concentration would be determined by the partial pressure of oxygen in the atmosphere. This means that the second boundary condition can be written as,

$$-D \frac{\partial C}{\partial x} \Big|_{\text{surface}} = K_3 (C_o - C_s), \quad (13)$$

where K_3 is a constant of the proportionality. To re-construct the dose rate effect, we must apply this surface resistance condition.

Consequently, the damage density, C_d , at the irradiation time of T , is obtainable using the following relation as:

$$C_d(x, T) = \int_0^T K_2 [P \bullet] C(x, T') dT'. \quad (14)$$

If the radical and oxygen concentrations reach steady state in early stage of the irradiation, this can be replaced by a simple multiplication. There are many parameters required in the present model. These were summarized in the literature (Yamauchi et al., 2003b).

As results of the calculations, the depth-distribution of the damage in the gamma-irradiated CR-39 was interpreted as a result of the exhaust of the oxygen in the deeper layer (Yamauchi et al., 2003b). The damage was formed only near the surface region where oxygen was supplied during the irradiation from air. The dose-rate effect on the bulk etch rate was also explained by introducing the surface resistant boundary condition for the feed of the oxygen from air. Almost equal reaction between the radical and the oxygen is expected to occur in the latent track formation process. The attained parameters for the present work would be useful to develop a new track formation model, in which the temporal and spatial dependence for the track was included. The previous experiments and the present calculations on the etching property of gamma-irradiated CR-39 would imply that the latent track in CR-39 was also produced

through the local radiation induced oxidation process along the ion path.

4. Vibration spectra

4.1. Introduction

It has been confirmed that CO₂ was produced in the CR-39 track detectors irradiated by various ions or low LET radiations (El-Shahawy et al., 1992; Chong et al., 1997; Saad et al., 2001; Malek et al., 1999, 2002). The ejection of CO₂ and CO gases from the bombarded CR-39 and the decrease of the density of C=O and C-O-C bonds in it have been also reported (Rickards et al., 1992; Gagnadre et al., 1993; Darraud et al., 1994; Gerstener et al., 1999). These observations suggest the scission of the carbonate ester bonds in the main chain of the CR-39 polymer during the exposure to radiation. However, the resultant product of the created damage, in other words, what is left after CO₂ release, is still not clear. On the other hand, OH group has been observed in the irradiated CR-39 (Yamauchi et al., 2000, 2001c). It is difficult to discriminate the absorption band of OH group from that of adsorbed water because of the hygroscopic property of CR-39 (Malek et al., 2000; Yamauchi et al., 2002d). Further studies are required to obtain information for the understanding of the latent track structure in CR-39. My colleagues and I have also made vibration spectral studies, FT-IR, Raman and Near-IR for CR-39 irradiated by various types of ions and gamma-ray (Yamauchi et al., 2000, 2002, 2003c). We are now trying to make a discriminative measurement, by setting-up of a FT-IR system on the beam line of the accelerator. A preliminary result is given in this conference (Yamauchi et al., 2002).

4.2 Formation of OH group

In Fig. 5, typical examples of IR spectra of gamma-irradiated CR-39 around OH absorption band are shown. The sample was irradiated with a dose of 100 kGy and then stored in vacuum below 10⁻⁵ Pa more than 1day. The spectra were obtained at times after the air leak. There are three peaks at 3470, 3550 and 3635 cm⁻¹, which are assigned to be the first overtone of C=O band, OH band and/or the symmetric vibration of water, and the anti-symmetric vibration of water, respectively (Gagnadre et al., 1993; Malek et al., 2000). As shown in these spectra, the absorption increases with the storage time in air and the saturated spectra are attained within 180 min. After the

measurements, the sample was out-gassed and the similar IR observation was made again. We have obtained almost equivalent spectra to that presented in Fig. 5. It was inferred that amount of water became was absorbed in the irradiated samples than that in unirradiated one. The created OH groups may be active-sites to retain the water from surroundings.

4.3 Overview for the track chemistry in CR-39

Chemical processes follow to the physical one. After irradiation, CO₂ gases are formed and the strength of C-O-C and C=O bands decrease apparently with the fluence. The absorption intensity of CO₂ gases was clearly dependent on the time after the air-leak, as well as the total fluence. More detailed measurements are required for the changes in the density of CH₂. The band of OH group was ascertained to increase in the height with the storage time. Some parts of this absorption were related to the absorbed water from the atmospheric humidity. On line FT-IR measurement showed that other parts would be associated with the creation of OH group. We have confirmed the important role of oxygen on forming damage related to the chemical etching, but the chemical reaction of the oxygen is not clear yet.

The CO₂ was formed as a result of the main chain of CR-39 breaking. The release of CO₂ gas would leave the damaged region with a lower density along the ion path. The observed OH group was also inferred to exist along the region, which could act as a water-absorber. The formation of CO₂ and OH group should be essential to understand the track formation process in CR-39 plastics, as well as that of the oxygen dissolved. The concentration of OH group would relate to the rate of the penetration of etching agent along the latent track. In the near future, the track etch rate in CR-39 will be described as a function of the concentration of OH group, as well as the physical stopping parameters.

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Figure Captions

Fig. 1. Fraction of area occupied by track cores as a function of the fluence at various track core radii.

Fig. 2. Fraction of area occupied by multiply overlapping tracks.

Fig. 3. Relation between the critical fluence and the track core radius (Yamauchi et al., 2003a).

Fig. 4. Damage distribution in depth and the bottle-like etched track shape.

Fig. 5. IR spectra of gamma-irradiated CR-39 around the band of OH group after different times of air-leak.

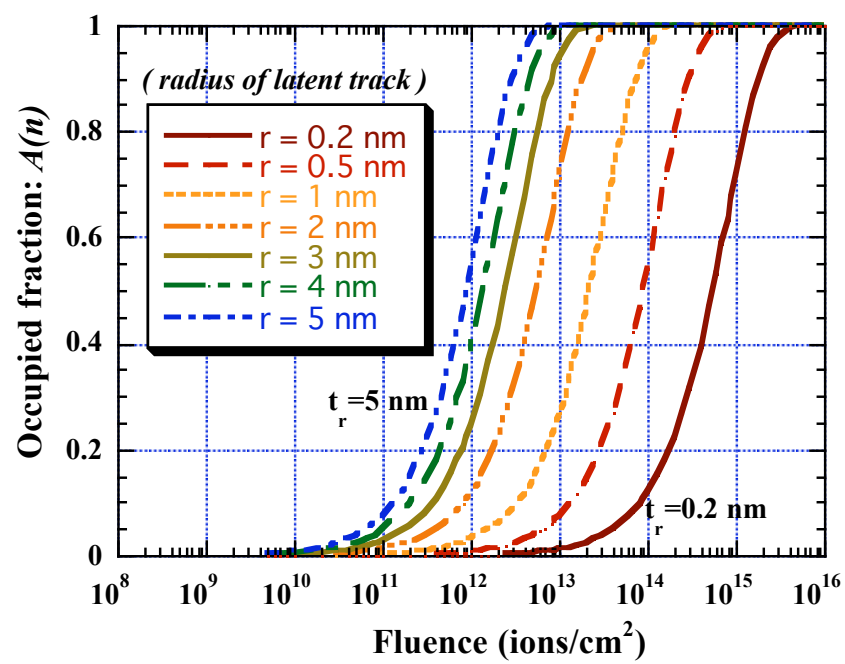


Fig. 1. Tomoya Yamauchi

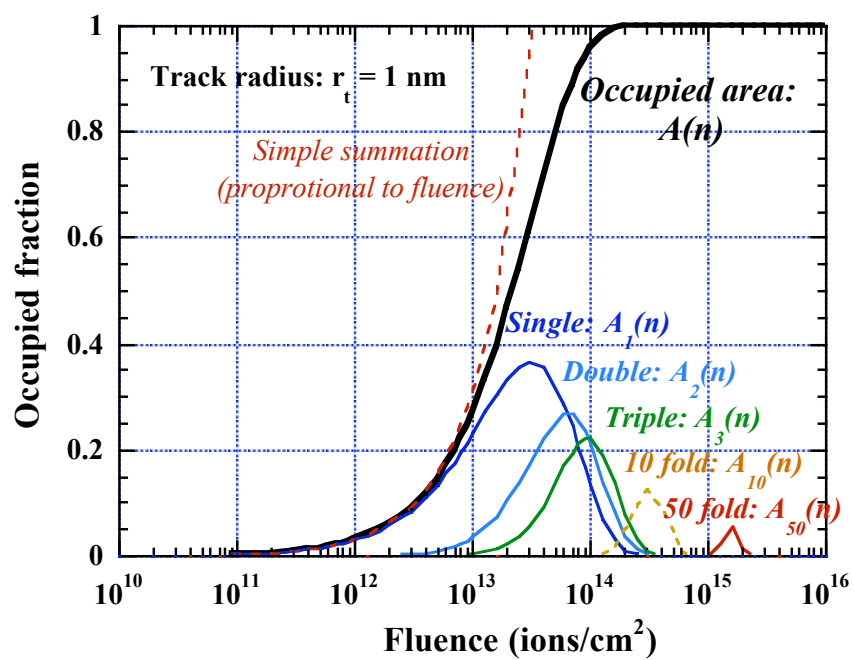


Fig. 2. Tomoya Yamauchi

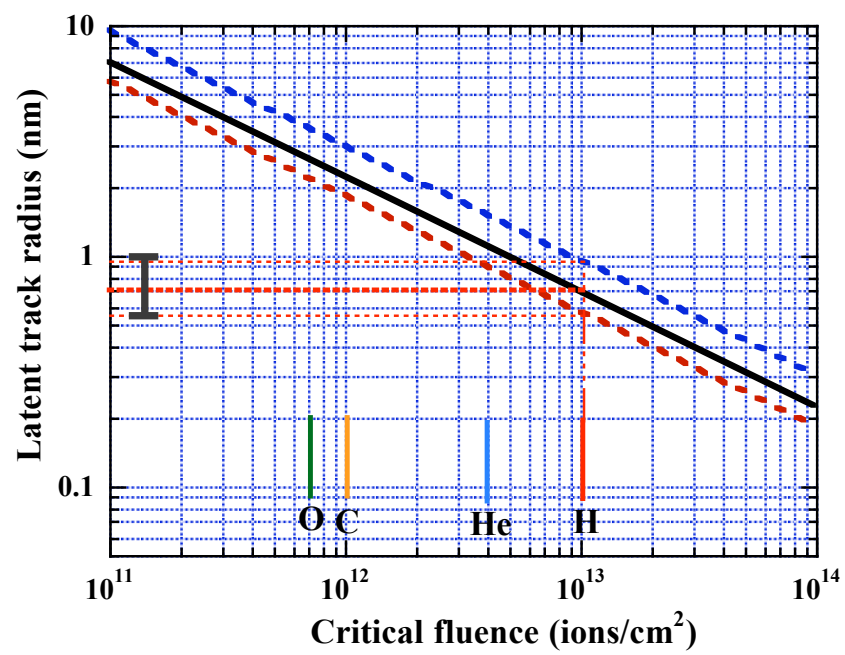


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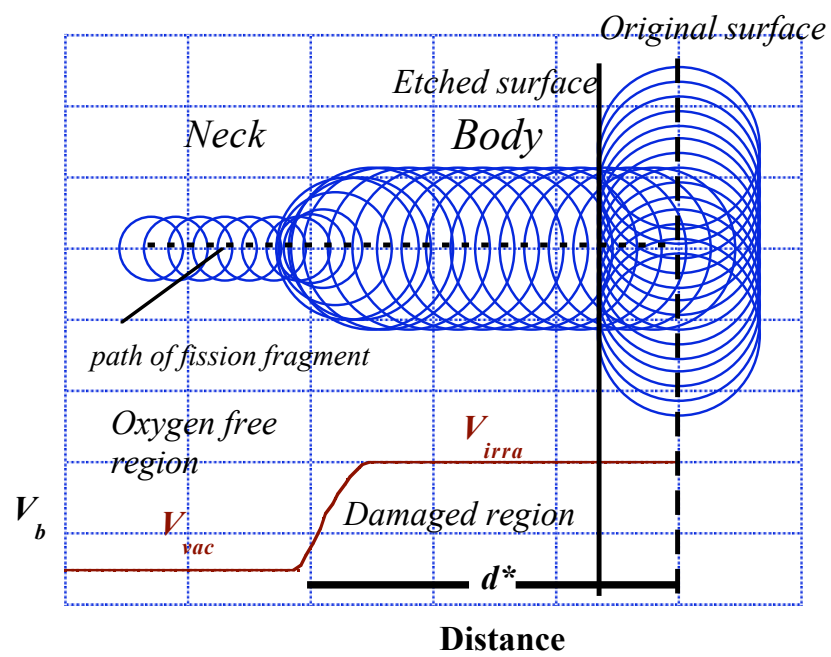


Fig. 4. Tomoya Yamauchi

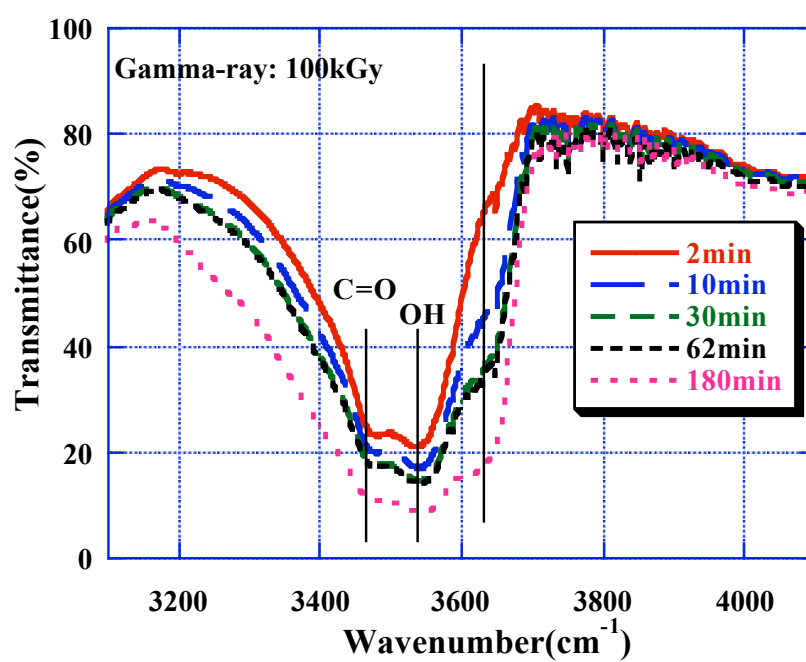


Fig. 5. Tomoya Yamauchi