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Track Distiribution of Recoil Protons in PN-3 Dosimeters Etched in NaOH Solution

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Abstrat. The method of etching tracks in PN-3 dosimeter has been applied to tracks of recoil protons from a neutron source. Both the etch and the detection response of PN-3 has been studied as a function of etched-track diameters against various parameters. We could obtain very useful informations about charge, energy, and mass of particles and the relationship between the track etching rate and the track forming procedure in order to analyze the prarticle recorded in the solid state track detector. The best etching condition could be found by means of changing the etching circumstances for various energies and particles in order to detect the charged particle accurately. It could be influenced widely that the polymer plastic detector could develop the detecting technique for the low energy level neutron and could be used as a neutron dosimeter in the radiation field such as the nuclear power station, the medical institute and the nondtructive testing institute.

I. Introduction

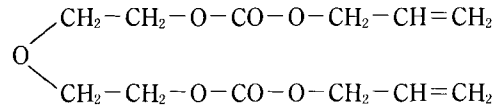
Since the introduction of allyl diglycol carbonate material (PN-3) as a solid state track recorder intense efforts have been made to confirm its properties and to develop its use in numerous fields of applications which are likely to be widespread in the fields of nuclear science and technology. Nuclear-track-recording solids can be used to identify energetic nuclear particles, because the size of the conical pit that can be develeped at the intersection of the particles trajectory with the surface is a function of the rate of energy loss of the particle. It has become known that the tracks of nuclear particles of different type and energy, registered in the same detector material, display a different behaviour in the course of the etching process.

The diameters of the etched tracks are specially sensitive to the parameters characteristic of the nuclear particles (energy and type of particles). The experimental results[1], recently reported, have justified the value and applicability of the information obtainable from the track diameters which vary during the etching process resulting from delta-ray generation[2] and ought to provide the greatest resolution of any charged particle detection system yet developed. In practice, non-uniformities and inhomogeneities in the track-recording solids now being used have prevented their theoretical resolution form being attained. However, the ideal polymer detector[2] should be an optically clear, amorphous, radiation-sensitive, thermoset material that does not cross-link upon irradiation and which is susceptible to interfacial degradation by a convenient etchant. One of the useful features of plastics is the possibility of interrupting and then undertaking the development again. This allows the option of measuring the tracks

during their formation, and of obtaining useful information for the particle identification (charge, energy and mass).

II. Chemical Property of PN-3 Track Dosimeter

PN-3 is polymerised from the liquid monomer, allyl diglycol carbonate, which has the following stucture[3]:



The present of the two allyl functional groups ($-CH_2-CH=CH_2$) in the monomer enables polymerisation to occur in a cross-linked network leading to a thermoset plastic. Characterising the extent of the most intensely damaged region of the core is an important step in understanding the nature of a particle track. The effect of radiation damage is primarily to lower the activation barrier for excising polymer molecules.

The usefulness of dielectric track detectors depends primarily on their ability[1] to :

1) Detect havily ionizing particles in a dense background of less ionizing radiation,
2) Detect rare heavy particles emitted at a rate less than one per year on a surface of a few cm^2,
3) Identify particles of a high energy-loss rate with a resolution higher than plastics that given by other detectors.

It is a fact that most tracks are ionization - produced defects - the result of the interaction of a charged particle with electrons attached to atoms in the detector. A fast, charged particle will eject electrons form atoms that were close to its path, giving them a distribution of energies that is strongly peaked toward low energies. Consequently, although the electrons carry a portion of the original energy of the incident particle to sites that are far from its path, most is concentrated only the path so that the residual effcets of the defects left or created where the electrons were removed are at, or near, the core of the track. Charecterising the extent of the most intensely damaged region of the core is an important step in understanding the nature of a particle track. The effect of radiation damage is primarily to lower the activation barrier for excising polymer molecules. A fraction of the energy dissipated by a charged particle is stored in the material and puts the damaged material into a higher energy state where it is more susceptible to attack.

The chemistry of track etching has been studied,[5, 6] and Paretzke *et al*[7] have examined the reaction between bisphenol-A polycarbonate (e. g. Lexan) and NaOH. The highly senstive track recording plastic CR-39, produced from allyl diglycol carbonate, has been studied.[5] They found that attack by the hydroxide ion results in the hydrolysis of the carbonate ester bonds and the release of poly-allylalcohol (PAA) from the polymer network. The reaction is[5]:

$$[CH_2=CH-CH_2-O-CO-O-CH_2-CH_2-O-CO-O-CH_2-CH=CH_2]+4OH^-$$
$$\rightarrow 2[CH_2\text{-}CH\text{-}CH_2\text{-}OH]+HO-CH_2-CH_2-O-CH_2-CH_2-OH+2CO_3^{2-}$$

In addition to the polymeric etch product PAA, 2'2'−oxydiethanol is also formed the above reaction.

The structure of a latent charged particle track is rather complex and difficult to deal with experimentally and theoretically. The same is true for the details of the chemical and physical process taking place when an etchant reacts with a latent track.

Since the discovery of the neutron, there have been various attempts to use it, especially based on the registration of proton tracks[9-11] has long been recognized as a basic requirement for a neutron detector.[12] This is one reason why PN-3 (or CR-39) is such a useful detector in this context. The proton response of it depends strongly on the etching conditions which are employed.[12] The probe method was taken in order to use PN-3 dosimeters as a track detector.

By far the most general, useful means of observing particle tracks in solids has been the technique of preferential chemical attack to enlarge and display tracks. The utility of the technique derives from its simplicity only common chemicals being needed and from the effective magnification that results from enlarging etched tracks to sizes where they can be viewed with an ordinary optical microscope. The procedure has allowed tracks to be observed in many dozens of substances.

There are two etching methods which are widely used the electrical etching[9, 12, 13] and the chemical etching. The electrical etching is simpler than the chemical etching. However, the chemical etching is used to enlarge the tracks in solids to visible size in general and the chemical etching method was chosen here because it allows better control over the etching process. Also it is well established that the electrical etching results in a loss of definition of track dimensions.

III. Experimental Procedure

1. Experimental Equipments

Neutrons can be detected indirectly by means of measuring photons or charged particles produced from the interaction between neutrons and nuclei. If the interaction system is know, the information of the interaction can be described by studying the reactions and the products of neutrons.

There are two interactions between the neutron and the nuclei : the absorption and the scattering. The absorption is (n, α), (n, p), (n, γ) or (n, fission). In general, the proton recoil method is used as the scattering of neutron-proton collision from which the detector can record the recoil protons.

Fig. 1 shows the geometry of the source and the moderator, and the positions in which the PN-3 dosimeters were irradiated. Fig. 1 (A) shows positions of each bar named from *A* to *E* according to its position from the centre where the 1.3 Ci Am/Be source was placed. Fig. 1 (B) shows the details of each bar which has four foil holders. The distances of each position from the source are:

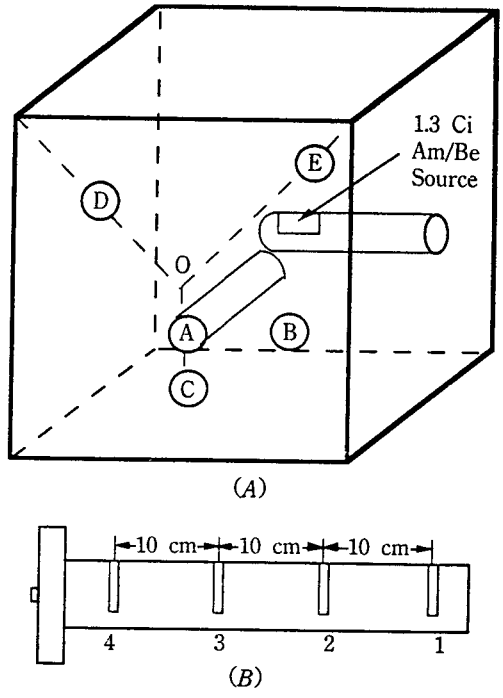


Fig. 1 The Schematic Diagram of the Moderator.
(A) Positions of the Neutron Source and Bars (A~E) in the Moderator.
(B) Slit Positions (1~4) in the Bar.

$A1$= 3.5 cm	$A2$=10.5 cm	$A3$=20.3 cm	$A4$=30.2 cm
$B1$= 8.5 cm	$B2$=13.1 cm	$B3$=21.7 cm	$B4$=31.1 cm
$C1$=19.0 cm	$C2$=21.5 cm	$C3$=27.6 cm	$C4$=33.5 cm
$D1$=25.5 cm	$D2$=27.4 cm	$D3$=32.4 cm	$D4$=39.4 cm
$E1$=38.0 cm	$E2$=39.3 cm	$E3$=42.9 cm	$E4$=48.4 cm

The notation of a position is Nn where N is the bar lockation (N=A~E) in the moderator and n is the position of the slit in the bar (n=1~4). For example, $A1$ means the nearest bar from the centre and from the source, i.e., $A1$ would be the nearest positon from the source and $E4$ was the farthest from the source.

2. *The Method of Neutron Track Detection*

Several PN-3 dosimeters were placed at different positions in a wax moderator for several days. After irradiaion, the dosimeters were pre-etched for two hours using a pre-etch solution of 60% methylated spirits plus 40% 6.25 Normal NaOH at 70℃ and the main-etched for six hours using 6.25 N NaOH soution at 70℃.

The greatest difficulty encountered in the process was the achieverment of a constant temperature over the eight-hour period. An electrically heated sand bath was used to heat a beaker of the solution which was magnetically stirred during the etching process. Even though the heater was thermostatically controlled, a problem was experienced in the control

of the solution temperature. Without a cover on the top of the beaker, the etching solution becoming more concentrated. This problem was partly overcome by covering the beaker and only heating the sand bath to the minimum temperature necessary. After the end of the etching process the etched PN-3 was rinsed using hot deionised water[9] at a temperature of about 80℃.

Six PN-3's were irradiated in the moderator at the same time and then etched together. After etching the dosimeters, the number and size of tracks per unit area were measured, having first calibrated the microscope graticule using a standard calibration slide. The PN-3 dosimeters were scanned using a ×10 eyepiece lens and ×45 objective lens, i. e., the total magnification of ×450 microscope in which each major graticule area was 1.6 μm×1.6 μm. This was subdivided into 100 equal squares. The length and diameter of a typical track was often smaller than the 10×10 graticule subdivisions and this gave to rise quite large errors.

Initially both the track diameters and their density over 6~9 different graticule areas were measured for each dosimeter. A background dosimeter, which had not been irradiated and which came from the same batch, was etched in the same solution and for the same time as each series of irradiated PN-3s. The track density in each irradiated dosimeter was calculated from each individual reading with an error Δt given by:

$$\Delta t = \sqrt{(\Delta m)^2 + (\Delta B)^2}$$

where Δm and ΔB are the errors in the measured and background values respectively.

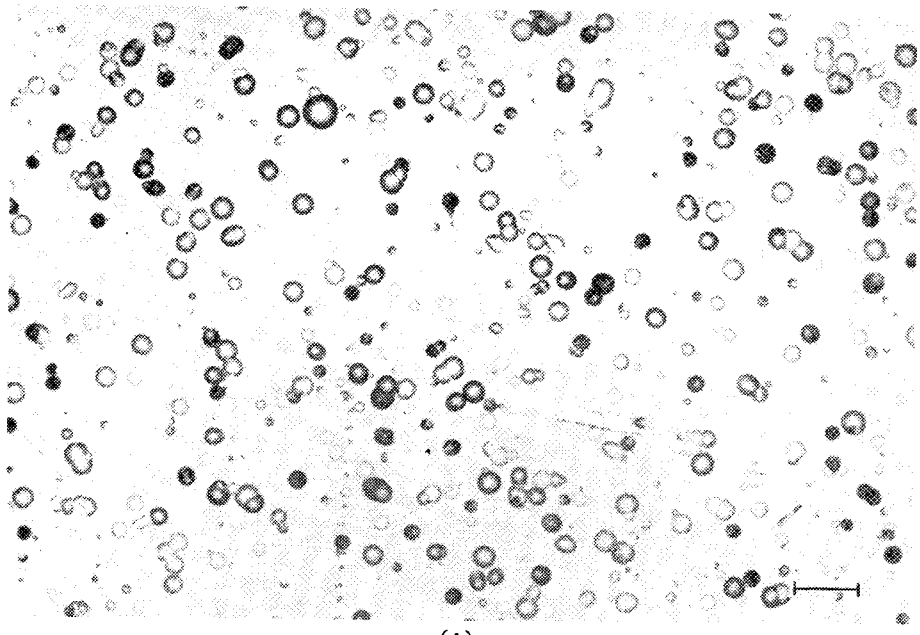
Bacause the neutron exposures in the moderator were rather short (~24 hours), the number of proton recoil tracks in this area was not large (0~15) and so many such areas on the PN-3 had to be scanned to obtain sufficient statics. The fluxes of "thermal" neutrons in each are given by the activities of the indium foils which were irradiated previously.

Ⅳ. Experimental Results

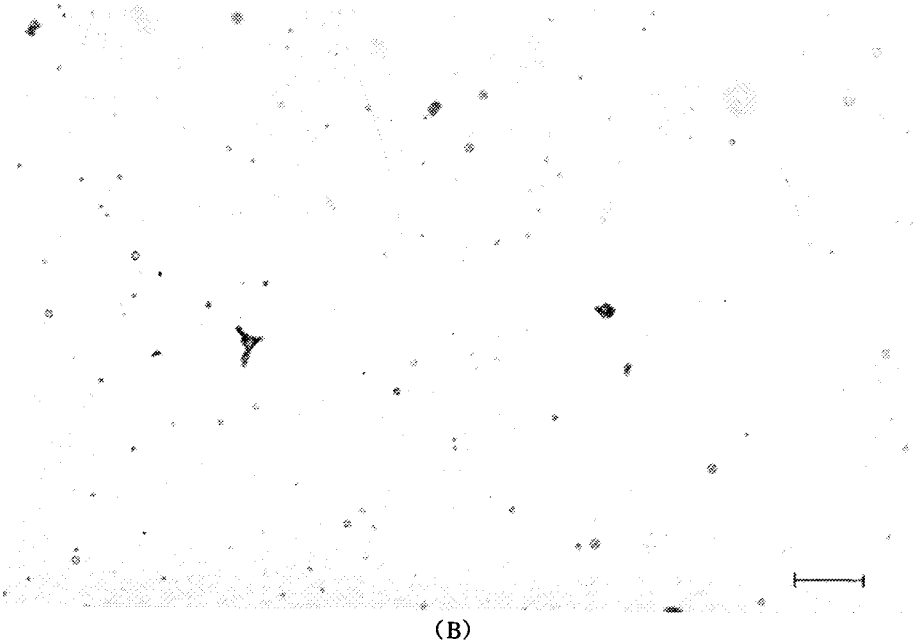
The results of neutron irradiation were presented as track density (i. e. number of tracks per unit area) in each irradiation position, for each track size (diameter). No information was obtained at this stage on track length.

Fig. 2 (A) shows the shape of the recoil proton tracks in the PN-3 dosimeter and (B) shows the background of those series of the dosimeters. The shape of tracks is almost circle (actually the cone). Fig. 3 shows how the distribution of track diameters varies with distance from the neutron source. Negative values of the track number in Fig. 3 are obtatined by subtacting track numbers of each position in the moderator from those of the background : the track number of background is denser than that of each position in the moderator at that track diameter. For the first exposed PN-3's, it can be seen that the number of large size (>1 unit) decreases as the distance from the neutron source is increased. A decrease of the number density of tracks of size 1 unit (=160nm) is clearly seen.

Fig. 4 shows the track distribution curves plotted as the track density against the distance from the source in the moderator. Positions of PN-3 dosimeters irradiated in the moderator

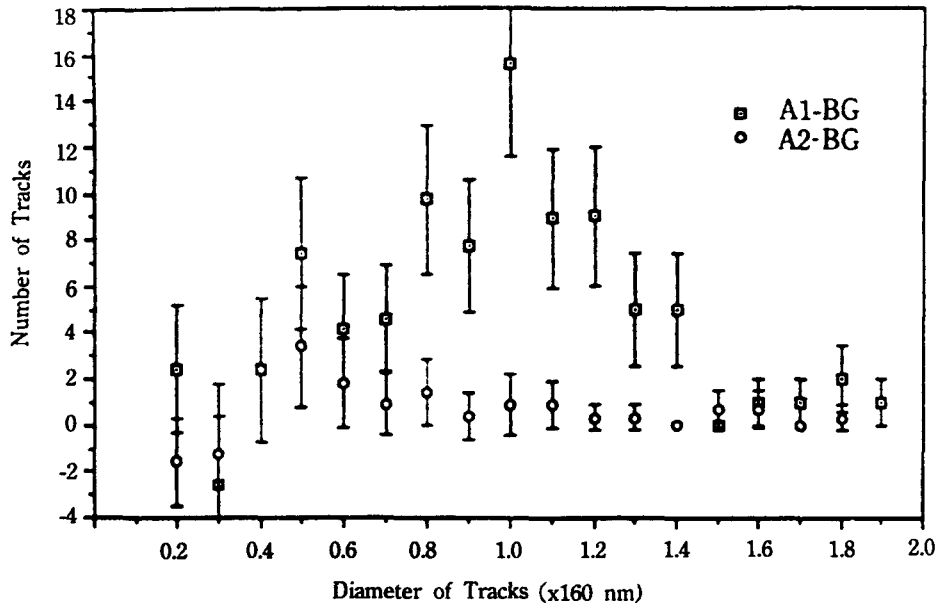


(A)

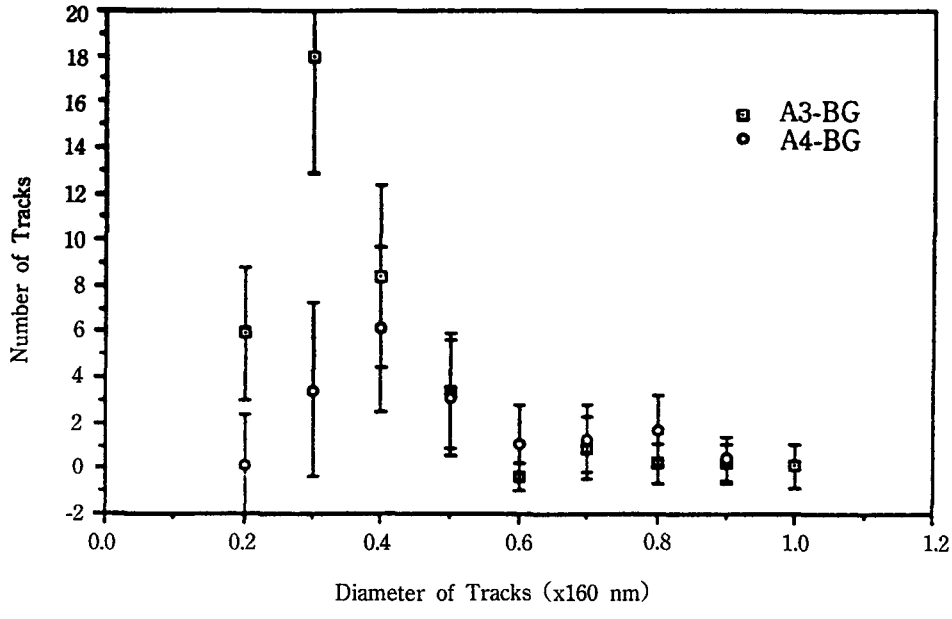


(B)

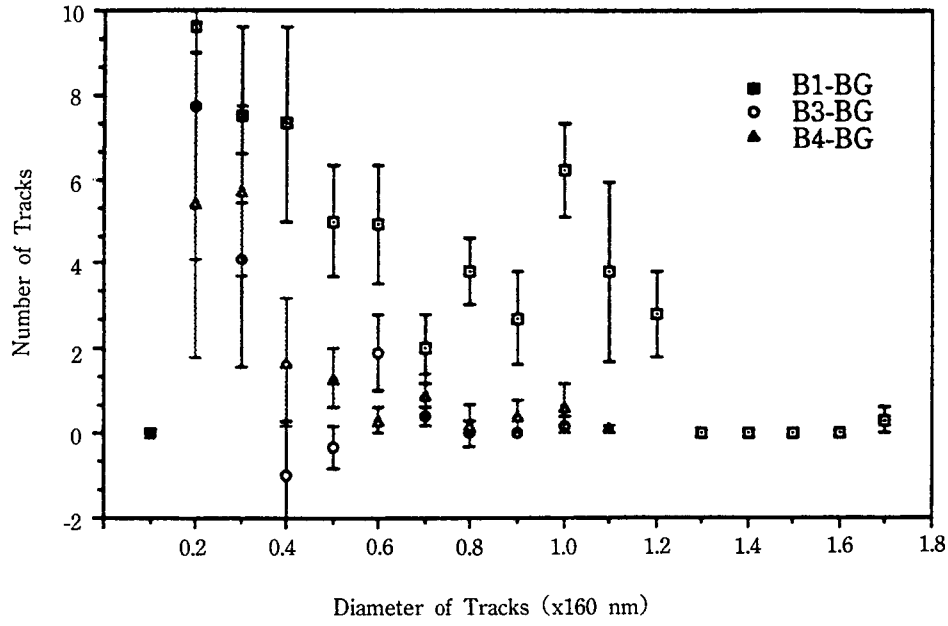
Fig. 2 Tracks in PN-3 under the ×100 Magnificaiton.
(A) Recoil Proton Tracks from the Neutron Source at *B1* in the Moderator and (B) Backgroud of PN-3 Dosimeters. The Scale is 0.5 μm.


A1-BG
A2-BG
Number of Tracks
Diameter of Tracks (x160 nm)

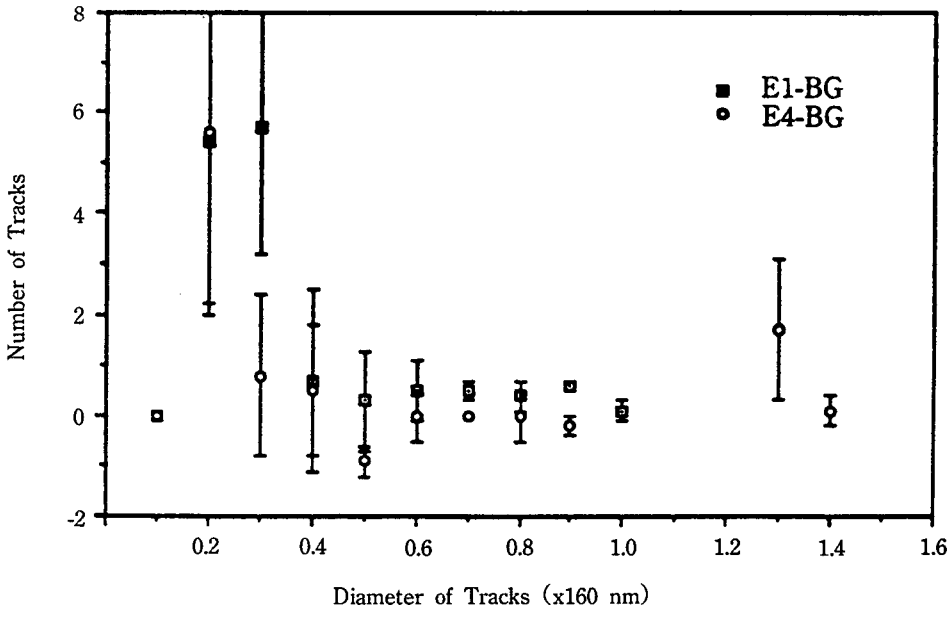
(A)


A3-BG
A4-BG
Number of Tracks
Diameter of Tracks (x160 nm)

(B)



(C)



(D)

Fig. 3 Distribution of Tracks at Each Position in the Moderator.
(A) and (B) Track Distributions on *A*, (C) on *B* and (D) on *E* in the Moderator.

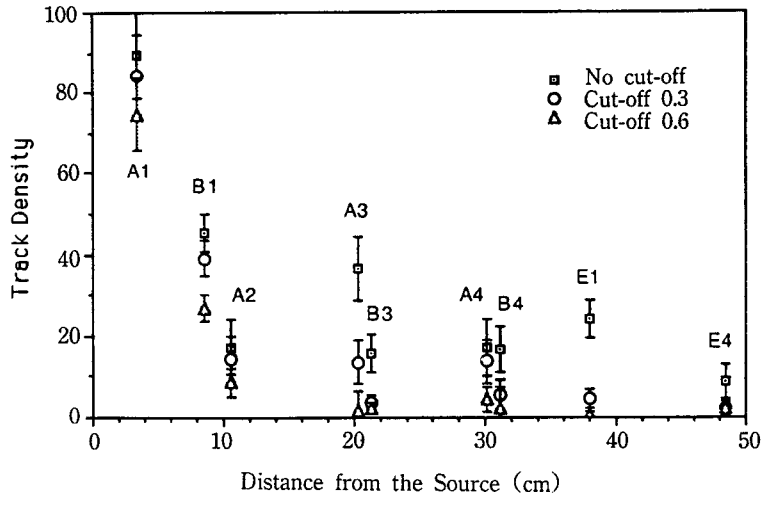


Fig. 4 Density of Tracks as a Function of the Distance from the Source Varying the Cut-Off Level.

are shown in the Figures (e. g. *A*1, *B*1 etc.). The data of the density in Fig. 4 were obtained by summing the track numbers from the arbitrary "cut-off" level (for example, cut-off 0.3 means cut off 0.3 unit and 1 unit = 160 nm) to the largest size of tracks in each graph in Fig. 3. The cut-off levels were chosen according to the distribution of each position in the moderator and to the distribution of the background. It can be seen that the track density decreases as the distance from the neutron source increases and that the distribution curve of the track density decreases smoothly after about 10 cm from the source in the moderator as the cut-off level is increased. The difficulty encountered in counting (or summing) the number of tracks in each curve was the large variation in track diameter (see Fig. 2 and Fig. 3).

V. Conclusions and Discussions

We could obtained very useful informations about charge, energy, and mass of particles and the relationship between the track etching rate and the track forming procedure in order to analyze the particle recorded in the solid state track detector. The best etching condition could be found by means of changing the etching circumstances for various energies and particles in order to detect the charged particle accurately. It is expected that these results could help the research for the field of Radiation Protection, Radiation Material and Medical Physics.

The method of etching tracks of PN-3 dosimeter has been applied to tracks of recoil protons from a neutron source. Both the etch and the detection response of PN-3 has been studied as a function of etched-track diameters against various parameters. For the neutron detection using PN-3, the distribution curves were plotted as a function of the track density (i. e., the number of tracks per the major graticle area) aganist the diameter of tracks. It

was difficult to find the general relationship between the distribution of tracks and the different position in the moderator. It can, howerver, be seen that the track density is decreased as increase of the distance form the source (see Fig. 4) and the distribution of tracks is likely to depend on the distance from the source because the distribution on $B4$ was consistent with the curve on $E1$ quite well (see C and D in Fig. 3).

The sensor material of PN-3 for the detection of neutron was high quality track-etch polymer manufactured by Vinten, England and the number of tracks is proportional to the exposed level could be controlled. It is possible to use PN-3 as a personal dosimeter and as a neutron detector in nuclear physics field. It could be influenced widely that the polymer plastic detector could develop the detecting technique for the low energy level neutron and could be used as a neutron dosimeter in the radiation field such as the nuclear power station, the medical institute and the nondestructive testing institute.

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NaOH 용액에 의해 부식된 PN-3 선량측정계에서의 되튕긴 양성자의 궤적 분포

유돈식, 심광숙
고려대학교 물리학과

중성자 선원에 의한 되튕긴 양성자의 궤적을 PN-3 검출기를 이용하여 화학적 부식방식으로 검출하는 방법에 대해 논의해 보았다. 또한 PN-3의 부식 및 검출특성을 부식된 궤적지름과 여러 변수들로 표시하여 보았다. 본 연구를 통해 고체 비적 검출기에 검출된 입자를 분석하기 위해 입자의 전하, 에너지, 질량과 궤적 부식 비율 결정 및 궤적 구조 형성 과정 사이의 관계식에 대한 자세한 정보를 얻을 수 있었다. 또한 하전입자를 보다 정확하게 검출하기 위해 입자의 종류 및 에너지에 대한 부식 조건을 변화시키면서 이에 대응하는 최적의 부식조건을 경험적으로 찾아내었다. 아울러 기대되는 기술 저변 확대 효과로는 고분자 플라스틱 검출기를 이용한 저준위 중성자 측정기술의 축적 및 개발을 통해 원자력발전소, 비파괴검사기관 및 의료기관 등 방사선 동위원소 취급기관의 중성자 선량측정계로의 유용한 응용 가능성이 예상된다.