Journal of NUCLEAR SCIENCE and TECHNOLOGY, Supplement 4, p. 514–517 (March 2004)

Deuterized Radiator for Detection of High-Energy Neutrons with Plastic Nuclear Track Detector

Keiji ODA[1,*], Yuichi IMASAKA[1], Tomoya YAMAUCHI[1], Yoshihiro NAKANE[2], Akira ENDO[2] and Yasuhiro YAMAGUCHI[2]

[1]*Department of Nuclear Engineering, Kobe University of Mercantile Marine, 5-1-1 Fukae-minamimachi, Higashinada-ku, Kobe 658-0022*

[2]*Tokai Establishment, Japan Atomic Energy Research Institute, Tokai-mura, Naka-gun, Ibaraki 319-1195*

(Received July 24, 2003 and accepted in revised form , 2003)

In order to develop a passive-type personal dosimeter based on plastic nuclear track detector (PNTD) for high-energy neutrons, a selection of radiator material and its thickness is one of key-points as well as utilization of high-sensitive type of PNTD. The radiator effect has been preliminarily estimated by numerical calculations using the angular differential cross section and characteristic data for charged particle detection with PNTD. A hydrogen compound, commonly used as a radiator, was expected to be not so effective for energetic neutrons higher than 10 MeV, so that a deuterized material was checked as one of the alternatives. The performance of these materials has been investigated through experiments in a quasi-monoenergetic neutron field, and the measured results were compared with those obtained numerically. It was also recognized that the dependence of relative sensitivity on the radiator thickness was considerable affected by a contamination of lower energy component in the neutron field.

KEYWORDS: ***plastic nuclear track detector, high-energy neutrons, radiator, deuterized material***

I. Introduction

In recent years, accelerated proton beam and secondary neutrons have been widely utilized for a variety of up-to-date applications to material science, environment science, life science, medical cancer therapy, *etc.* A great number of accelerator facilities have already been in steady operation, and more are in construction or planning in many countries. In Japan, a collaborative big project of JAERI (Japan Atomic Energy Research Institute) and KEK (High Energy Accelerator Research Organization) is now in progress. Such a proton beam eventually generates high-energy neutrons in slowing down or stopping in a material. These neutrons are harder to be detected than lower ones, because the efficiency of the existing monitors and dosimeters becomes lower. Hence, it is very important to monitor them correctly, in other words, to establish a countermeasure for efficient detection of high-energy neutrons for radiation protection purpose.[1)]

The detectors in actual radiation protection dosimetry are classified into two categories, which respond to the dose rate at a working place and the personal dose to a worker, respectively. The fixed area monitors and portable survey-meters belong to one group of active detectors. The other is personal dosimeters, which are further subdivided into real-time response type and accumulation type according to the length of measurement time period. For X- or γ-rays, for instance, a pocket-type semiconductor detector and a glass dosimeter (radio-photo-luminescence) are typical of two groups of personal dosimeters. For neutrons with energies lower than 14MeV, a semiconductor dosimeter and plastic nuclear track detector (PNTD) replace them, respectively.

It has already been pointed out that the prevailing PNTD such as CR-39 detector has less sensitivity to high-energy neutrons because of a difficulty in registering energetic proton recoils.[2-4)] The use of the detector without any corrections would bring about a serious underestimate of the radiation dose and dose rate. Thus, the authors started to develop an advance technique for more efficient detection of high-energy neutrons. The experimental studies require a well-defined and clean neutron source, such as TIARA (Takasaki, Ion Accelerator for Advanced Radiation Application) facility, JAERI.

In order to increase the detection efficiency, it is necessary to make two major improvements; one is to find a most sensitive PNTD element, and the other to sensitize it with an appropriate radiator supplying charged particles to PNTD.

In the previous experiments[5-7)], we checked the performance of three types of PNTDs; "BARYOTRAK" of pure CR-39 plastic, "TD-1" of an improved-type CR-39 and "TNF-1" of a co-polymer based on CR-39, which were made by Fukuvi Chemical Co., Ltd., Japan. From a comparison among them in relative efficiency for 65-MeV neutron detection and in optical property after chemical etching, "TD-1" was chosen as a promising element, which is CR-39 plastic containing a small amount of anti-oxydant. As to the radiator, we investigated the effect of four types of radiator materials; polyethylene (CH_2), deuterized dotriacontane ($C_{32}D_{66}$), lithium fluoride (LiF) and graphite (C). It was found that the increased sensitivity of the deuterized material exceeded that of CH_2 for a relatively thin radiator of the order of a few hundreds of mg/cm^2, and that

*Corresponding author, Tel. & Fax. +81-78-431-6304,
E-mail: oda@cc.kshosen.ac.jp

other materials of LiF and C worked unsatisfactorily.

We proceed the study on high-energy neutron dosimetry to the next step. It is the purpose of this report to confirm the effect for a radiator thicker than g/cm^2, comparable with the maximum range of recoil particles. A theoretical approach was also tried to support the experimental results.

II. Numerical Calculations of Radiator Effects

1. Calculation Model

The etch-pit formation in PNTD exposed to fast neutrons takes place in two steps; production of secondary charged particles through neutron interactions with constituent atoms in PNTD and radiator materials, and visualization of their latent tracks by the chemical etching. The overall detection efficiency is the sum of two components of the intrinsic efficiency and the enhancement by the radiator, which are distinguished by the position of charged particle production, *i.e.* inside or outside the PNTD. It is well known that the latter dominates for neutron energies higher than several hundreds of keV.[2-4] The radiator effect, increase in the efficiency by a radiator, is defined here as the number of charged particles per incident neutron, which are generated in the radiator, reach PNTD and are observed as etch-pits. It is expressed by the following formula of double integral of the interaction yield with respect to the solid angle, Ω and the depth in the radiator, z:

$$\varepsilon = N\iint \frac{d\sigma}{d\Omega}\, d\Omega\, dz \quad , \tag{1}$$

where N is the number density of atomic nuclei interacting with neutron and σ is the cross section for the interaction.

The integral range in eq. (1) should be limited by two major conditions characteristic of chemically etched PNTDs. One is so-called critical angle condition, by which inclined latent tracks are eliminated. The other is related with a recognition of small etch-pits with an optical microscope and image processing system, which reflects on the reduction of z-integral range.

2. Procedure of Numerical Calculations

The following assumptions were made in the preliminary calculations;

1) contribution of particles heavier than proton or deuteron is ignored because of their short range,
2) neither attenuation of incident neutron nor multiple scattering in the radiator is, for simplicity, taken into account, and
3) the etch rate ratio, an important parameter of response of PNTD to charged particles, is independent of the angle of incline.

The energy-dependent cross sections were referred to the nuclear data file, ENDF/B-VI of newest version. The differential cross sections, given in the files as a function of neutron scattering angle in the center of mass system, are converted into those with respect to the angle of recoil particles in the laboratory system.

For a given angle, all the latent tracks can not be observed as etch-pits because of both critical-angle and pit-size conditions described above. **Figure 1** represents upper and lower energies, which was calculated by using the response data of TD-1 detector to protons reported by Ogura et al.[8] The particle tracks between the cut-off energies can be countable as etch-pits. Then, z-integral in eq. (1) is replaced by the difference between the ranges corresponding to both energies.

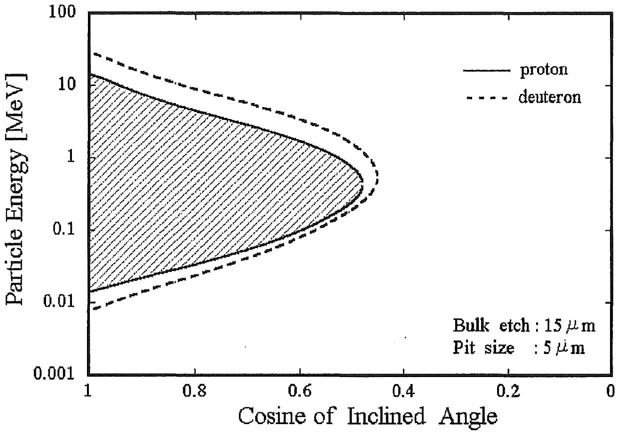

Fig. 1 Proton and deuteron energies observed as etch-pits after 15 μm etching.

3. Results of Numerical Calculations

The results of numerical calculations of eq. (1) are summarized in **Fig. 2**, where enhancement in detection efficiency is shown as a function of radiator thickness for several neutron energies. A hydrocarbon such as polyethylene is most popular as a radiator for fast neutrons, but a gelled water is assumed in this calculation by experimental reasons stated in the succeeding section. It is recognized that the effect saturates near the thickness comparable with the proton range of the maximum recoil energy, and that a relatively thick radiator of the order of g/cm^2 is required for several tens of MeV. In general, it becomes more difficult for PNTD to catch proton recoils as the neutron energy increases. The kinematics suggests another candidate of massive particle, deuteron. A dotted

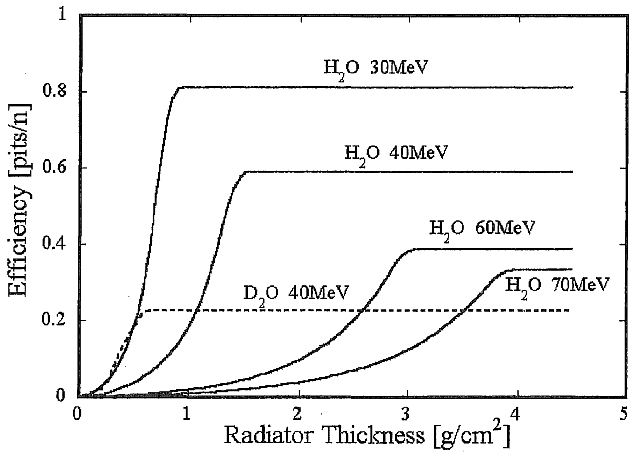

Fig. 2 Dependence of the increase in detection efficiency upon the radiator thickness

line in **Fig. 2** represents the results for 40-MeV neutrons for a heavy water (D_2O) radiator. The saturation value for D_2O radiator does not exceed that for H_2O, but is attained by thinner radiator owing to short range of deuteron recoils.

The saturated value, which is considered to be an ideal case, is plotted against the neutron energy in **Fig. 3** for respective radiators. The hydrogen radiator is still more effective for all the energies except near 30 MeV. It is found from **Figs. 1** and **2** that the superiority between two materials should depend on both the neutron energy and the radiator thickness. The fact suggests a technique for control of the energy dependence by a combination of two types of radiator materials. Such an attempt is now in progress and will be stated in near future.[3)]

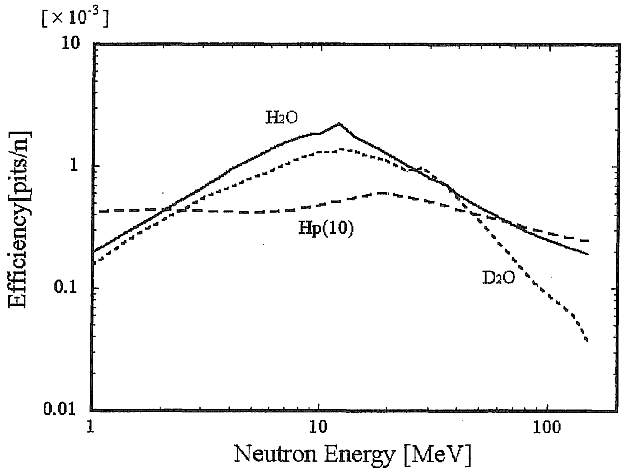

Fig. 3 Radiator effect as a function of the neutron energy.

III. Experimental Results and Discussion

A neutron irradiation experiment was carried out at TIARA, JAERI, where quasi-monoenergetic neutrons were generated by bombarding a lithium target with an accelerated proton beam. These neutrons were guided to an irradiation room through 220-cm thick iron collimator[9)], and then hit TD-1 detectors.

We searched a deuterized material appropriate for radiator. A dotriacontane, $C_{32}D_{66}$ is one of candidates but too expensive for preparing a thick radiator of the order of g/cm^2. So, we utilized heavy water in this experiment, which was gelled by adding 1% agarose.

TD-1 detectors were then chemically etched in a stirred 7M NaOH solution at 70 °C. The number of etch-pits was counted with a semi-automated system consisting of an optical microscope, image processing system and personal computer.

The results were summarized in **Fig. 4**, where the incremental etch-pit density was plotted against the radiator thickness. For thinner radiators below 2 g/cm^2, D_2O radiator dominates H_2O as was predicted by numerical calculations. The measured dependence on the thickness, however, is a convex shape differently from a concave pattern for monoenergetic neutrons shown in **Fig. 2**.

One of major reasons of this discrepancy is considered to be a contamination of lower energy neutrons in a quasi-monoenergetic field. An exact energy spectrum has already been measured by Baba et al.,[9)] as is shown by a dotted line in **Fig. 5**. The distribution was divided, for simplicity, into several parts shown by a solid line, and the contribution of each component was summed up by weighting the fractional portion. **Figure 6** shows the result obtained in this procedure, together with that for mono-energetic neutron field. It is found that the increase pattern is sensitively affected by lower energy neutrons.

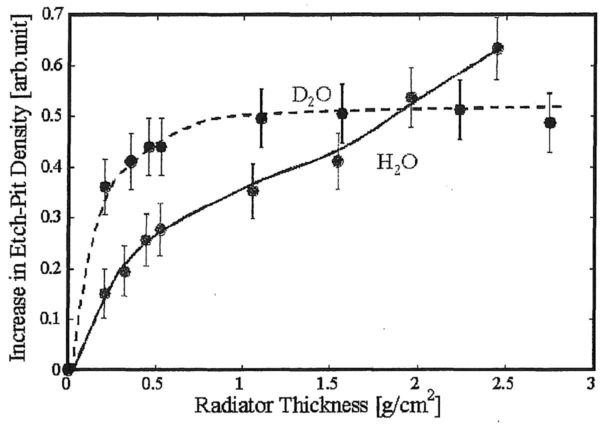

Fig. 4 Measured etch-pit density as a function of the radiator thickness for 65-MeV quasi-monoenergetic neutrons.

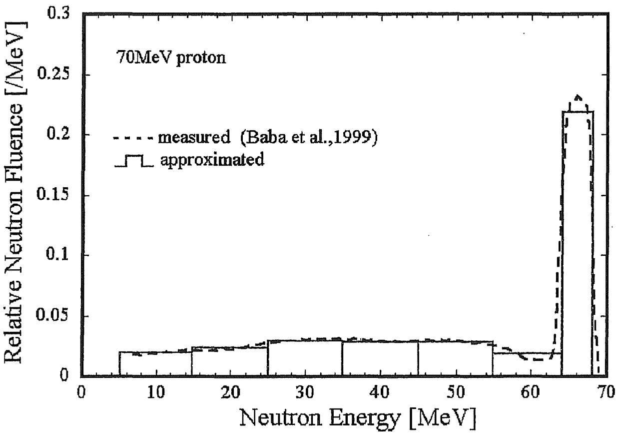

Fig. 5 Energy spectrum of quasi-monoenergetic neutrons measured by Baba et al. [9)]

As was described above, the response of PNTD with a thick radiator to quasi-monoenegetic neutrons has been discussed both theoretically and experimentally. The thickness, however, is one of the critical problems for practical applications to personal dosimetry; namely it is appropriate to reduce the weight of the radiator for convenience. Another important problem still remains to adjust the energy dependence of the overall sensitivity to that of the conversion factor for radiation protection purpose shown as $H_p(10)$ in **Fig. 3**. We expect a two-layer type radiator[2)] is one of promising techniques to control the energy dependence.

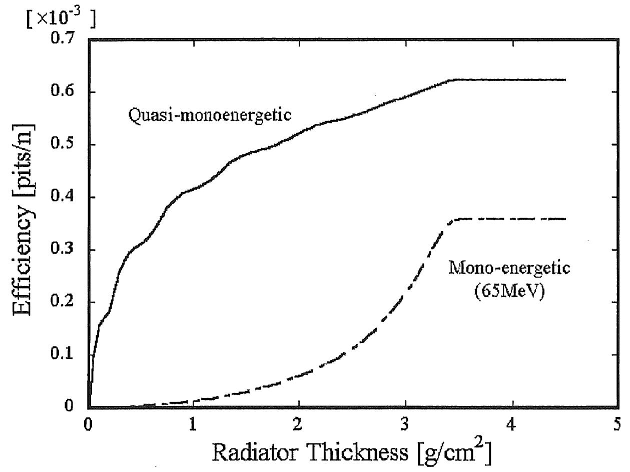

Fig. 6 Radiator effect for monoenergetic and quasi-monoenergetic neutrons.

IV. Conclusion

As part of a study on personal dosimetry of high-energy neutrons, a sensitization by radiators was discussed. The increase in the sensitivity has been obtained experimentally for a thick radiator of the order of a few g/cm^2. It was confirmed that the thickness dependence of the radiator effect was modified sensitively by contamination of lower energy neutrons. The efficiency has also been calculated under several simplified conditions. The results showed that the effect of deuterized material should exceed that of hydrogen radiator for neutrons of about 30 MeV. Such a deuterized material or a combination of two materials is expected to be a special radiator for high-energy neutrons.

Acknowledgements

The authors would like to express their sincere thanks to all the staffs of TIARA facility, especially Dr. S. Tanaka, for their help of neutron irradiation experiments. We are also indebted to Mr. K. Tsukahara, Kobe University of Mercantile Marine for his assistance for experimental arrangement.

This work was undertaken as part of the special project research between JAERI and universities in Japan, and partially supported by Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology.

References

1) Y. Yamaguchi and H. Hirayama, "JAERI-Universities Joint Research Project on Radiation Safety in Proton Accelerator Facilities," to be published in this journal.
2) K. Oda, M. Ito, H. Yoneda, H. Miyake, J. Yamamoto amd T. Tsuruta, "Dose-equivalent response CR-39 track detector for personnel neutron dosimetry," *Nucl. Instrum. Meth. Phys. Res.*, **B61** [2], 302-308 (1991).
3) D. Hermsdorf, R. Bretschneider, B. Dorschel and J. Henniger, "Neutron response calculation on the basic of variable track etch rates along the secondary particle trajectories in CR-39," *Radiat. Meas.*, **31** [1-6], 431-436 (1999).
4) R. J. Tanner, D. T. Bartlett and L. G. Hager, "Recent enhancement to the understanding of the response of the NRPB neutron personal dosemeter," *Radiat. Meas.*, **34** [1-6], 457-461 (2001).
5) K. Oda, H. Ichijo, N. Miyawaki, T. Yamauchi and Y. Nakane, "Improvement of neutron detection efficiency with high sensitive CR-39 track detector," *Radiat. Meas.*, **34** [1-6], 171-175 (2001).
6) K. Oda, Y. Saito, N. Miyawaki, T. Yamauchi, A. El-Rahmany, Y. Nakane and Y. Yamaguchi, "Characteristic response of plastic track detectors to 40-80 MeV neutrons," *Radiat. Prot. Dosim.*, **101** [1-4], 569-572 (2002).
7) K. Oda, Y. Imasaka, K. Tsukahara, T. Yamauchi, Y. Nakane and Y. Yamaguchi, "Radiator effect on plastic nuclear track detectors for high-energy neutrons," *Radiat. Meas.*, in press (2003).
8) K. Ogura, M. Asano, N. Yasuda and M. Yoshida, "Properties of TNF-1 track etch detector," *Nucl. Instrum. Meth. Phys. Res.*, **B185** [1-4], 222-227 (2001).
9) M. Baba, Y. Nauchi, T. Iwasaki, T. Kiyosumi, S. Yoshioka, N. Hirakawa, T. Nakamura, Su. Tanaka, S. Meigo, H. Nakashoma, Sh. Tanaka, and N. Nakao, "Characterization of a 40-90 MeV ^{7}Li(p,n) neutron source at TIARA using a proton recoil telescope and a TOF method," *Nucl. Instrum. Meth. Phys. Res.*, **A428** [2], 454-465 (1999).