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Several techniques for improving energy dependence of a commercial personal neutron dosemeter package based on PADC track detector

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Abstract

A practical applicability of several techniques to a commercial dose-assessment service has been investigated for improving the energy dependence of the personal neutron dosemeter based on PADC plastic track detector. Under a constraint of no modification in detector material and the fixed chemical processes, two technical attempts have been investigated; an analysis of etch-pit data obtained by microscopic observation and a devise of radiator structure. In addition to the number of etch-pits, another information of their distribution in some parameter has been obtained in the former technique. It was found that the two-window method was possibly applicable to the distribution in the etch-pit size and the gray level. In the latter, a performance of radiator-degrader structure has been confirmed experimentally, and it was found to be one of most promising techniques.

Keywords: Neutron personal dosemeter; Energy dependence; Etch-pit distribution; Two-window method; Radiator-degrader

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1. Introduction

Since Cartwright et al. have discovered the excellent property of proton registration of Poly-allyl diglycol carbonate (PADC, CR-39) plastic (Cartwright et al., 1978), a number of applications of PADC to fast neutron detection have been reported by many authors (Benton et al., 1980; Hooton et al., 1982; Turner et al., 1984; Oda et al., 1991a; Spurny, 1995; Tanner et al., 2005). They are roughly classified, from a methodological point of view, into an improvement in PADC manufacture, optimization of etching condition, sensitization by radiator, experimental study of response to monoenergetic neutrons, theoretical approach to prediction of absolute sensitivity, and so on. On the contrast to the activities of these sophisticated techniques, the applications of PADC detectors to routine personal neutron dosimetry in actual workplaces is very few. In this report, our concern is directed from scientifically challenging subjects to practical application to dose assessment on commercial base. In this situation, more attention should be paid to stability in all processes, accuracy in dose-assessment and speed in etch-pit analyzing system.

Chiyoda Technol Co., Ltd. (CTC) is the biggest company for radiation protection dosimetry service in Japan. Its dosemeter package consists of PADC, a polyethylene sheet of 1 mm thick as a radiator for fast neutrons and a boron nitride sheet for thermal neutron detection. In order to secure the quality of detector elements, PADC plastic is manufactured in their own laboratory (Ohguchi et al., 2008a). In addition, a pre-soaking process is adopted because a considerable number of false pits are often observed on a post-etched surface of non-irradiated PADC sample, which results in serious reduction in the neutron sensitivity (Ohguchi et al., 2008b). A fast scanning microscope, HSP-1000 (SEIKO Precision Co., Ltd, Japan) is employed in the track counting process to handle more than 30,000 pieces per month. This dosimetry system in CTC is working well, but there remains, for higher quality service, an improvement in the energy dependence, especially about 40 % drop in the sensitivity at 15 MeV.

It is the purpose of this report to investigate an applicability of several techniques proposed in the literatures for increasing the relative sensitivity to 15 MeV neutrons, without any modification of PADC characteristics or any change in chemical processes. Under such conditions, we have two options; one is careful observation of microscopic pictures, by which additional information can be obtained such as distribution in the etch-pit size, gray-level and shape factor. In the other techniques, the structure of the radiator is modified in order to control the energy dependence.

2. Experimentals

The PADC plastic was manufactured in the laboratory in CTC to keep the characteristics of detector elements. The sheet of 1.6 mm thick was cut into small pieces of 19x9 mm.

An neutron irradiation experiment was carried our at monoenergetic fields of 0.14, 0.57, 5.0 and 15 MeV neutrons, generated by a Van de Graaff accelerator at National Institute of Advanced Industrial Science and Technology, Tsukuba, Japan.

In order to reduce the number of false pits, a pre-soaking technique was employed with a solution of Di-n-Butyl Phthalene (DBT). The conditions of the concentration, temperature of solution, duration of process were already optimized (Ohguchi et al., 2008b), and the contamination of false pits was reduced to be about one-tenth.

The PADC samples processed by the pre-soaking and chemical etching in a KOH solution were analyzed by a fast scanning microscopy system, HSP-1000 (Yasuda et al., 2005). It is composed of an optical microscope, a line sensor, three-dimensional stage and an auto-focus system, by which 48 mm^2 area of PADC sample can be scanned within one minute.

3. Etch-pit distributions in the parameters

As was reported in the previous paper (Ohguchi et al., 2008b), a good response of the dosimeter package could be experimentally achieved for 0.14, 0.57 and 5.0 MeV neutrons. We are going to discuss several techniques for compensating 40 % drop in the sensitivity to 15 MeV neutrons. In is necessary to derive an additional parameter other than the etch-pit number. In this section, we tried to find an energy-dependent variation in the etch-pit distribution in three different parameters.

At first, a few thousands of counted etch-pits were analyzed with respect to the diameter, as has been done by several authors (Turner et al., 1994; d'Errico et al., 1997). The result is shown in Fig. 1, where the normalized distribution for 15 MeV of interest is represented by a bold solid line. After careful observation, we recognized some difference in the region of the diameter longer than 25 μm. The contribution of these larger etch-pits amount to 5.3 % for 15 MeV neutrons, whereas 1 % or lower for other lower neutron energies.

The fact of relatively large component suggests an applicability of the two-window technique, which is often adopted in pulse-height distributions measured by active-type counters. In our case, a corrected sensitivity, S is calculated by a linear sum of two components divided by a discrimination level;

$$S = S_1 + aS_2, \quad (1)$$

where S_1 and S_2 is the partial sensitivity due to the etch-pits of which diameter are shorter and larger than the discrimination level of 26 μm, respectively, and a is the weighting factor. The corrected sensitivities to 5 and 15 MeV neutrons are shown in Fig. 2 by a broken and solid line, respectively, as a function of the weighting factor. Two curves intersect with each other at a factor of 14. The result implies that the correction by the two-window technique seems to be successful.

Another consideration, however, is indispensable for practical application; that is a statistical error. Surely there found a difference in relative abundance of larger etch-pits, but its absolute value of 5.3 % is very small. The statistical error in counting larger etch-pits is exaggerated by the weighting procedure according to the error propagation principle. Assuming 1 mSv exposure, as an example, the width of error bar is represented by dotted lines in Fig. 2. The relative error of 7 % in the sensitivity to 15 MeV neutrons becomes doubled by a weighting factor of 14. It is concluded from the discussion that the correction by a diameter cut-off is possible but involved unwillingly a significant degradation in precision.

Most researchers with an experience of microscopic observation of etch-pits induced by fast neutrons may know a difference in darkness of etch-pits between protons and heavy charged particles. So, we tried to analyze the gray-level of etch-pits, which defined here as a relative value of the average darkness on a microscopic picture. The distribution in the gray-level is shown in Fig. 3. Comparing among four data, we can find a two peaks only for 15 MeV neutrons. Up to now, we can not yet understand the reason why two peaks are formed in the gray-level distribution. Despite unclear theoretical confirmation, this characteristic peak in the gray-level between 105 and 120 may be experimentally utilized for correction for energy dependence. When the two-window method is applied, the weighting factor of 4 is found to be suitable. It is considered to be practical from a statistical point of view.

The etch-pit data are transformed into the distribution in the third parameter of the shape factor, defined here as the ratio of minor to major radius. It is recognized from Fig. 4 that a separation of four curves by this parameter is very difficult.

4. Design of radiator structure

Only polyethylene sheet of 1 mm thick is used in the present CTC dosimeter package. In this section, our concern is focused on another option of designing a radiator structure.

The deficiency in the neutron sensitivity in high-energy region may be caused not only the decrease in neutron cross section but also difficulty of energetic proton registration in PADC plastic. In such a high-energy neutron field, a partial polyethylene layer near the boundary does not work as a radiator, because the energy of the protons recoiled there and entering into PADC is too high to be recorded as a visible etch-pit. Thus, the authors have proposed a two-layer structure consisting of a deuterized hydro-carbon (CD_2) and a polyethylene sheet (CH_2) for high-energy neutrons (Oda et al, 2003; Oda et al., 2005). The CD_2 layer plays a role of both a radiator of deuterons produced there and a degrader for energetic protons recoiled in outer CH_2 layer. An optimum thickness can be estimated to be 60 mg/cm2 of CD_2 and about 200 mg/cm^2 of CH_2 for 15 MeV neutrons.

However, a deuterized material, for example, dotriacontane ($C_{32}D_{66}$) is so expensive for a commercial service, and too weak in mechanical strength to be practically applied to the commercial dosimeter package.

Matiullah et al. (1987) proposed a technique for reducing arbitrarily the neutron detection efficiency by inserting intentionally a thin radiator between PADC and the radiator. The authors have also tried to make different energy dependences by designing the thickness of both a polyethylene radiator and an aluminum degrader, and pointed out that the neutron energy spectrum could be roughly estimated by unfolding the responses of plural detectors (Oda et al., 1991b).

Thus, a preliminary experiment with an aluminum sheet of 0.4 mm thick was performed in the reference neutron fields. The result is shown in Fig. 5, where data for the normal dosemeter package and those with a radiator-degrader structure are represented by open and closed circles, respectively. Little change between two points is found for 0.14 and 0.57 MeV neutrons, which means most of charged particles forming countable etch-pits are produced within PADC plastics. On the other hand, the sensitivity to 5 MeV neutrons is drastically reduced by introducing a degrader, which means most of etch-pits are due to protons recoiled in a radiator.

The reason for increase for 15 MeV neutrons by using a degrader is closely related with a polyethylene thickness shorter than the range of 15 MeV protons, and the energy dependence of proton registration efficiency. The radiator thickness of 1 mm in CTC dosemeter package is sufficient for 5 MeV neutrons, but insufficient for 15 MeV neutrons supplying the protons with a maximum range of about 2.5 mm. As is explained above, a part of polyethylene near the boundary does not work well as a radiator but as a degrader for energetic protons produced in outer region of polyethylene. In other words, the effective thickness of the radiator is much shorter than the actual thickness of 1mm polyethylene. If an aluminum degrader is inserted, the radiator becomes substantially thicker and more effective for etch-pit formation.

The variation of the sensitivity in the region of several MeV or higher can be reproduced by numerical calculations taking into account an assumed response function for proton registration, the etch-pit formation criteria for the dip angle and the diameter (Oda et al., 2003). Figure 6 shows the calculated result of the radiator effect, i.e. the enhancement in the neutron sensitivity by a radiator, where a cross of two curves is seen at 15 MeV or higher.

The results shown in Figs. 5 and 6 suggest that the dependence of the sensitivity on the neutron energy may be controlled by combination of the radiator and the degrader. We are now trying to find an optimum thickness and in the next step to establish a technique of three-layer structure for better response in a wider energy region.

5. Conclusion

In this report, several techniques for controlling the energy dependence of personal neutron dosemeter have been investigated from a viewpoint of routine commercial service of dose assessment. The results obtained are summarized as follows:

1) Distribution in the etch-pit diameter is found to be an energy-dependent parameter, but may be difficult for practical application because of relatively large statistical error.

2) Distribution in the gray-level may be utilized for the two window method, but a theoretical consideration and an experimental confirmation for other neutron energies are required.

3) Distribution in the shape factor, the ratio of minor to major radius, has not changed distinctively with the neutron energy.

4) Deuterized radiator proposed in the previous report is possible to apply to the present dosemeter package, but commercially negative because of very high cost and weak mechanical strength of a deuterized material.

5) Radiator-degrader technique is one of promising methods for compensating the energy dependence. The thickness of both degrader and radiator is most important factor for successful structure of the dosemeter package. Additional experiments at other neutron energies and a theoretical prediction are required.

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Figure captions

Fig. 1. Normalized distribution in the etch-pit diameter.

Fig. 2. Corrected sensitivities to 5 and 15 MeV neutrons.

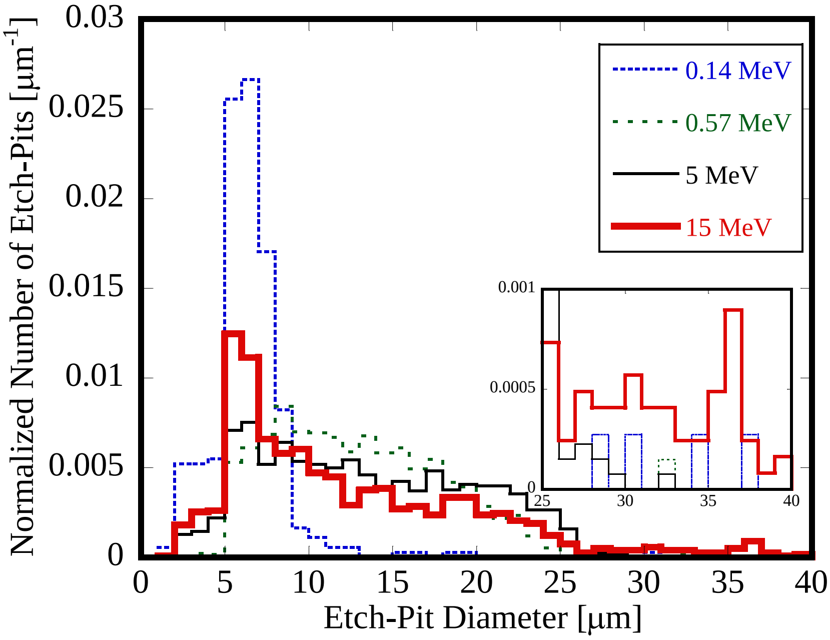
Fig. 3. Normalized distribution in the etch-pit diameter.

Fig. 4. Normalized distribution in the ratio of minor to major radius.

Fig. 5. Comparison of neutron sensitivity between radiator only and radiator-degrader structure.

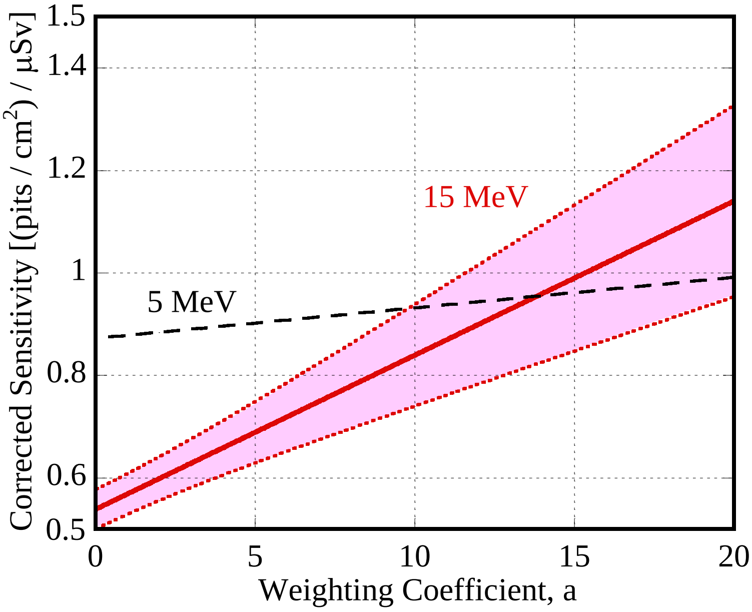
Fig. 6. Calculated radiator effect for polyethylene only and radiator-degrader structure.

Fig. 1



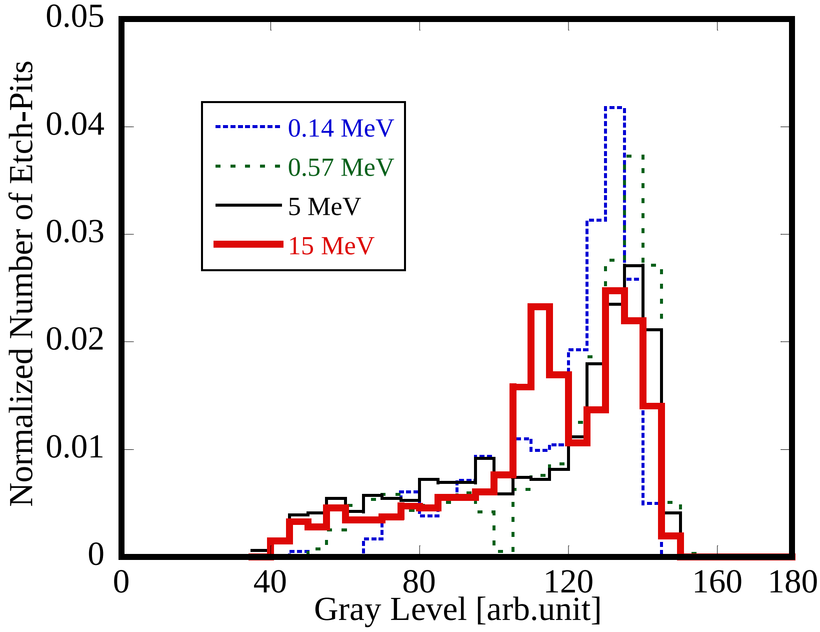
0.14 MeV
0.57 MeV
5 MeV
15 MeV
Normalized Number of Etch-Pits [μm^{-1}]
Etch-Pit Diameter [μm]
0.03
0.025
0.02
0.015
0.01
0.005
0
0
5
10
15
20
25
30
35
40
0.001
0.0005
0
25
30
35
40

Fig.2



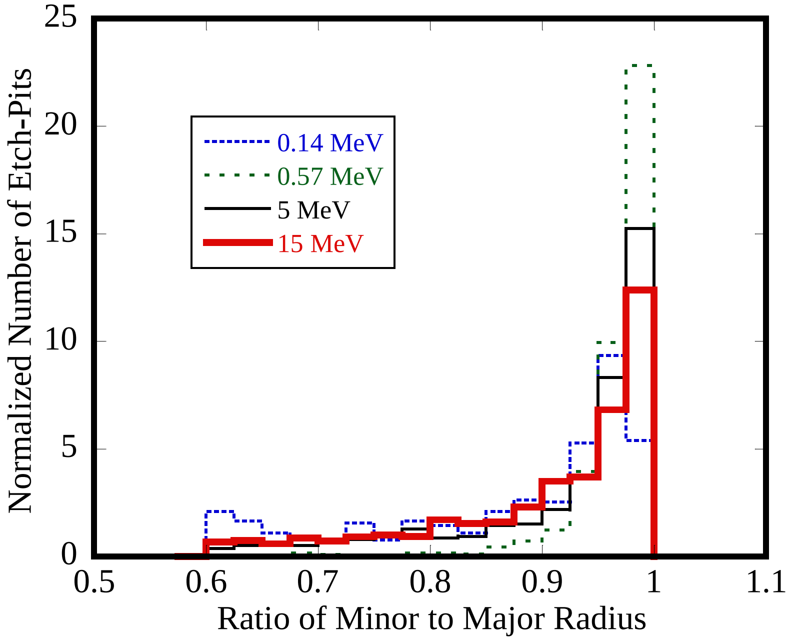
Corrected Sensitivity [(pits / cm2) / μSv]
1.5
1.4
1.2
1
0.8
0.6
0.5
15 MeV
5 MeV
0
5
10
15
20
Weighting Coefficient, a

Fig. 3



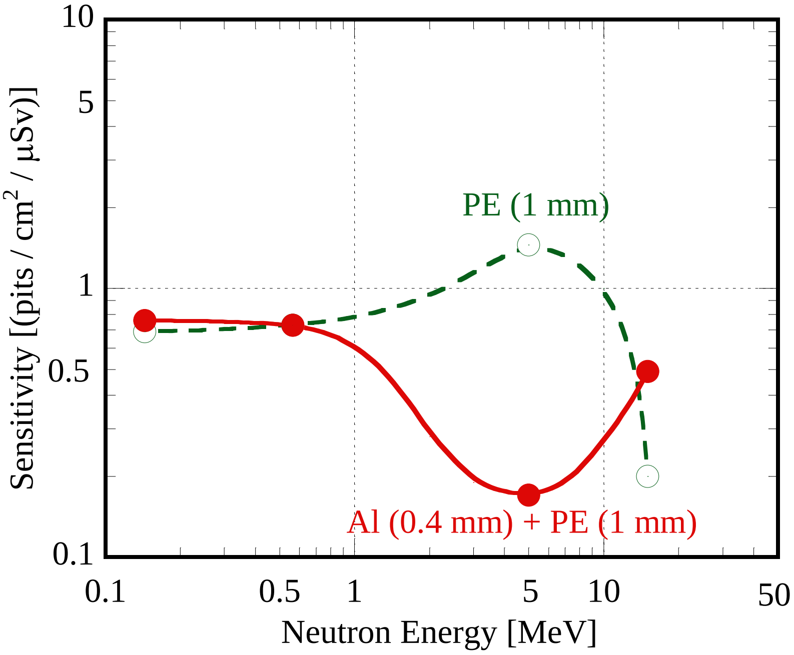
0.05
0.04
0.03
0.02
0.01
0
Normalized Number of Etch-Pits
0.14 MeV
0.57 MeV
5 MeV
15 MeV
0
40
80
120
160
180
Gray Level [arb.unit]

Fig. 4



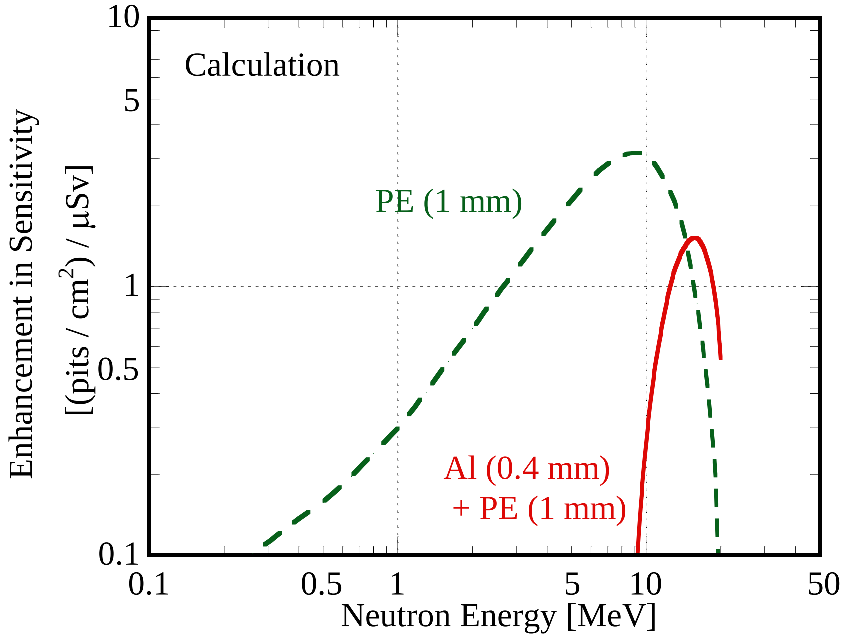
0.14 MeV
0.57 MeV
5 MeV
15 MeV
Normalized Number of Etch-Pits
Ratio of Minor to Major Radius
0
5
10
15
20
25
0.5
0.6
0.7
0.8
0.9
1
1.1

Fig. 5



10
5
1
0.5
0.1
Sensitivity [(pits / cm2 / μSv)]
PE (1 mm)
Al (0.4 mm) + PE (1 mm)
0.1
0.5
1
5
10
50
Neutron Energy [MeV]

Fig.6



Calculation
PE (1 mm)
Al (0.4 mm)
+ PE (1 mm)
10
5
1
0.5
0.1
0.1
0.5
1
5
10
50
Neutron Energy [MeV]
Enhancement in Sensitivity
[(pits / cm2) / μSv]