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Energy and Angular Response of CR-39 Neutron Track Detector

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중성자 비적 검출기 CR-39의 에너지 및 입사각 응답특성

김장렬 · 하정우 · 윤여창
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Abstract

Published data of the efficiency of CR-39 detectors as a function of neutron energy from different laboratories show wide variations in the response obtained. These variations result from differences in etching conditions, in the materials and thickness of the radiator, in the sensitivities of CR-39 from different manufacturers and perhaps in criteria used for the size of spots that are counted. This paper describes some effects of these factors on the energy and angular response with calculational results. Calculated and measured results of the variations of response with neutron incident angle are more consistent than those of energy response. The data calculated show that the angular response is not a strong function of neutron energy except below about 0.3 MeV.

요 약

중성자 에너지의 함수로써 여러 연구기관들에 의하여 발표된 CR-39검출기의 효율측정은 서로 많은 차이가 있음을 보여주고 있다. 이러한 차이는 부식조건, 방사체의 물질과 두께, 각 회사 제품에 따른 CR-39의 감도 그리고 계수하는 부식직경의 크기 등에 기인된다. 본 논문에서는 계산결과와 함께 에너지 및 각에 따른 분포에 대한 효과들을 검토하였다. 몇가지 에너지에 대해서만 수행된 중성자 입사각에 따른 반응변화의 계산 및 실험결과는 에너지 반응의 결과보다 더욱 일치하는 것을 알 수 있었다. 계산결과에 따르면 입사각에 대한 반응은 약 0.3 MeV 이하를 제외 하고는 중성자 에너지에 따라 심하게 변하지 않는다는 것을 보여준다.

1. Introduction

Since the early day of nuclear power generation two systems have been available for monitoring individual neutron doses. Both are based on photographic film. Thermal neutron doses can be monitored by the standard film dosemeter which contains a cadmium insert and fast neutron doses by nuclear emulsion dosemeter which contains a relatively thick nuclear emulsion which after development contains images of neutron-induced proton tracks. But the outstanding weakness of the dosimetry system constitute by these two detectors is that it cannot detect the neutrons between 1 eV and 700 keV which is very important range in working areas of power stations[1,2]. Also the nuclear emulsion dosemeter suffers from other disadvantages, it is tedious and tiring to evaluate and is prone to fading.

In the late 1960s the development of thermoluminescence dosimetry techniques led to the widespread introduction of albedo dosimetry systems based on the detection of thermal neutrons with materials such as thermoluminescent LiF-6[3,4]. These have advantages over the normal film badges in that the detection element is small and sensitive, and have a roughly constant dose equivalent response from thermal neutrons to about 20 keV. there has, however, remained a need to improve fast neutron dosimetry techniques and there has been a continuing search for better detection systems.

The first materials to be considered were polycarbonate which could be used to detect fission fragments and heavy recoils of carbon and oxygen from appropriate neutron converter materials. The most promising converter material was neptunium-237. This interacts with thermal, intermediate and fast neutrons to give easily detectable fission fragments, but is unfortunately expensive and significantly radioactive.[5].

A breakthrough occurred in 1978 when it was recognized that a polymer of diethylen glycol dis (allyl carbonate) generally called CR-39, a material developed in the 1930's, could be used to detect protons[6]. CR-39 is a hard clear plastic which is commonly used to make eye glass lenses and is readily available from a number of suppliers. The low energy threshold for proton is between 100 to 200 keV in the early use of CR-39[7] and with development may be even lower.

This paper describes the basic principles of CR-39 neutron damage track detector briefly and the experimental results of the neutron energy and angular response of this detector with calculational results.

2. Theory

1) Response of CR-39 to Protons

The majority of tracks produced by neutrons in CR-39 are from recoil protons, generated partly in the surface layer of the CR-39 that is removed by etching and partly in some hydrogeneous radiator in front of the CR-39. In Figure 1 the dashed curves show the calculated contributions of these two components for a particular set of conditions. At low energies recoils from the etched layer dominate, because recoil proton ranges are so short that the thickness of the layer of radiator from which protons can reach the CR-39 is less than the thickness of the etched layer itself. For high neutron energies, provided that the radiator is thick and rich in hydrogen, protons generated in a large volume of radiator can reach in the CR-39 and these recoils are much more numerous than those from the etched layers.

Calculations of the number of recoil protons that enter the etched layer or are generated in it, and their distributions in energy and angle, are very accurate. They depend on the (n,p)cross section, the angular and energy distributions of proton recoils and the range-energy relations for protons in the radiator material, all of which are accurately known for the energy range of 10 keV

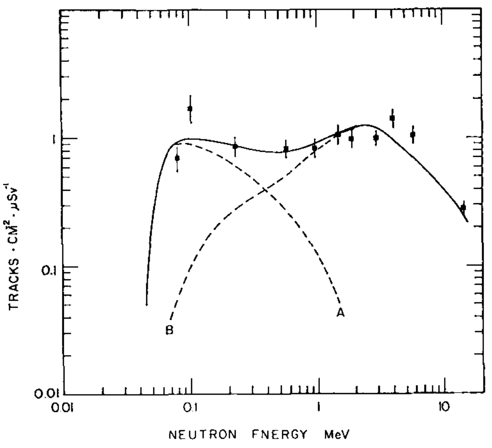

Fig. 1. Comparison of the Measured Response of a CR-39 Neutron Dosemeter with that Calculated (Solid Curve). Curve A shows the Contribution of Protons Generated in the Etched Layer While Curve B Gives the Contribution of Protons Produced in a Thick Polyethylene Radiator. From Cross et al.[8].

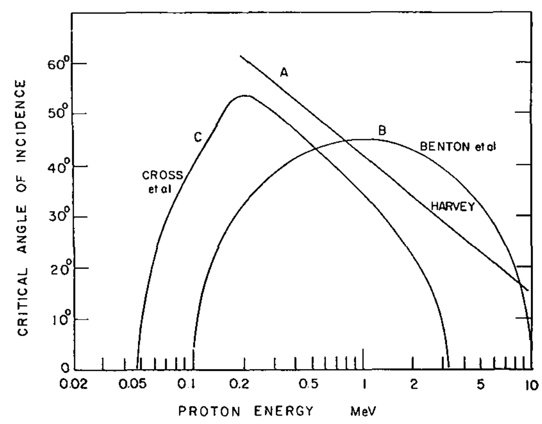

Fig. 2. Critical Angle of Incidence for Monoenergetic Protons in CR-39. The Data from References 8, 9, and 10.

to 15 MeV. What is not so accurately known is the fraction of these protons that give detectable tracks in CR-39 because of the limits required on their directions and energies. For chemical etching, any charged particle produces a visible track only if the angle between its direction and the surface normal is less than a critical value θ c, given by

$$\sin \theta c = V_B/V_T$$

where V_B and V_T are the bulk etching rate and the etching rate along a track, respectively. The latter varies with the particle energy, so the relation between the critical angle and the energy of protons has been determined for some type of CR-39 and under certain etching conditions.

Figure 2 shows the results of some of these measurements. Curve A is for Pershore CR-39, curve B is for American Acrylics CR-39 manufactured around 1980. Both curves were obtained using only chemical etching. Curve C applies to 1984 American Acrylics CR-39 processed by electrochemical etching. This comparison indicates that critical angles are smaller for electrochemical etching than for chemical etching. This is probably ture but has been proven only for one set of etching conditions.

All these curves must go to zero at some upper and lower energy. For electrochemical etching the minimum detectable energy depends on the electric field strength. At 30 kV/cm the minimum energy is about 50 keV as shown in figure 2: at 45 kV/cm can detect protons down to 10 or 20 keV.

The response to high-energy normally-incident protons cuts off when the etchant does not penetrate far enough along the track to give a local high field strength and produce electrical breakdown. In Curve C of Figure 2 the maximum energy of detectable protons is about 3 MeV. The electrochemical etching techniques typically used appear to only detect protons with energies up to 1 to 3 MeV. Protons of considerably higher energies could be detected with longer etching.

2) Calculation of Response to Neutrons

Provided that the relation between critical angle and proton energy is known, it is straightforward to calculate the number of proton tracks per incident neutron or tracks. cm^{-2} Sv^{-1} and this has been done by a number of investigators[8-11]. The

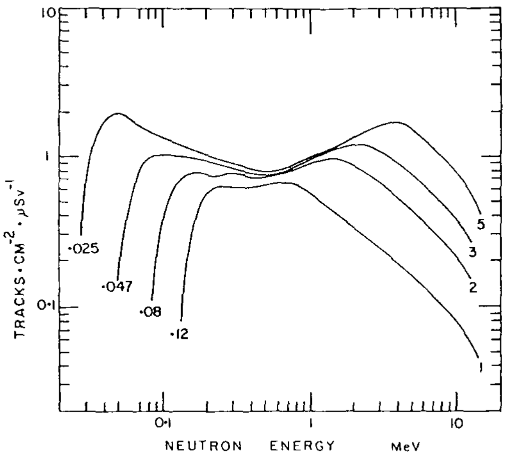

Fig. 3. Calculated Response of CR-39 Dosemeters for Various Maximum and Minimum Detectable Proton Energies. The Numbers on the Curves Give These Energies in MeV.

present calculations are described in reference 10. It is primarily just a matter of calculating proton ranges and evaluating an integral. We characterize the etching technique and type of CR-39 by three parameters—the minimum proton energy, which depends mainly on the electric field strength, the maximum energy which is expected to depend on the amount of etching, and the maximum critical angle which depends mainly on the type of CR-39, and assume that the shape of the curve relating critical angle to energy can be estimated well enough by comparison with the measured curve of Figure 2. The limits on proton energy and the value of the maximum critical angle are much more important to the shape of the neutron response than is the precise shape of the curve of Fgure 2.

Some examples of calculated responses are shown in Figure 3. The different curves apply to various upper and lower detectable proton energies, given on the curves in MeV, and all apply to "thick" polyethylene radiators. The upper ends of these response curves thus depend on the total amount of etching; the lower ends mainly on the electric field strength. The neutron energy at which the high-energy peak appears in these responses is just below the maximum detectable proton energy and the higher this energy is the greater is the height of the peak. The flattest response is given by a maximum proton energy of 2 or 3 MeV.

The general shape of these curves is easily understood. The number of tracks arising from neutron interactions in a thick radiator increases with energy because the proton range, which determines the volume of radiator from which protons can be collected, goes up faster than the (n,p) cross section, which determines the number generated per unit volume, goes down. The recoil protons are distributed in energy from zero up to the neutron energy. When the neutron energy increases above the maximum detectable proton energy a decreasing fraction of the protons reaching the CR-39 can produce visible tracks and the response goes down again. It is noteworthy that above the high energy peak in the response (and for a thick radiator) the slopes of the response curves are always the same, being exactly proportional to the (n,p) cross section. This is easily shown theoretically but will not be discussed here. It is only true if virtually all the response is from recoil protons.

The lower end of the response curve is fairly flat down to about 1.5 time the minimum proton energy and then falls rapidly to zero. The shape in this region is also affected somewhat by the thickness of the etched layer, which depends on the total amount of etching.

Figure 1 shows how the response measured with monoenergetic neutron agrees with that calculated. In this case the maximum and minimum proton energies and the critical angle for the etching conditions used are all known from measurements with monoenergetic protons, and no adjustments are made to match the calculated and measured values. If these quantities are not

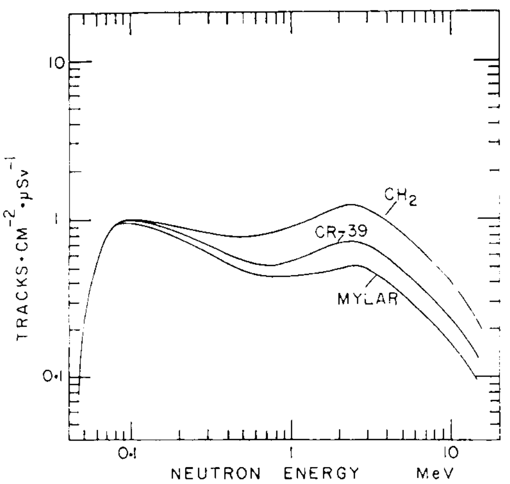

Fig. 4. Variation of the Energy Response of CR-39 Neutron Dosemeters with the Materials of the (Thick) Radiator

known they can be regarded as parameters and varied to match a measured response curve with calculated result.

3. Experiment

1) Descriptions of Dosemeters and Irradiations

The detectors were cut from a sheet of "dosimeter grade" (93% monomer purity) CR-39 manufactured by American Acrylics in 1986. Each sheet is covered by a protective layer of polyethylene, about 12 mg/cm^2 thick. Prior to irradiation the CR-39 was stored in air, in darkness and at room temperautre. Detectors were 0.63 mm thick and approximately 2cm $\times$ 2cm.

2.8 and 14.7 MeV monoenergetic neutrons were obtained from the neutron generator made by AECL at the Chalk River Nuclear Laboratories. Nuetrons of 2.8 MeV are from D(D, n)^{3}He reaction and those of 14.7 MeV from D(T,n)^{4}He reaction. To get the variation with radiator (CH_2) thickness of the response of the CR-39 dosemeter to 14.7 MeV neutrons, additional polyethylene radiators were placed in front of each detector, so the total thickness of polyethylene were from 0 to 250 mg/cm^2. The angles used to get the angular response were 0 to 90 by increasing 15 and dose equivalent was 400 mrem at each angle.

2) Processing and Assessment

Following the method of Tommasino et al.[12] we used electrochemical etching at 60°C. Etching was in 6N KOH for 5 hours, with a 60 Hz AC electric strength of 30 kV/cm (RMS). Before electrochemical etching, the CR-39 was chemically etched for 1 hour at 60°C to reduce the background and left it water-saturated air at 60°C for over night (about 16 hours).

After etching, tracks were magnified 76 times in a microfiche reader (3M model 297-BM) and counted by eye. About 0.4 cm^2 area was counted. The minimum diameter of counted tracks was about 0.5 mm.

4. Results and Discussions

1) Effects of Radiator Composition and Thickenss

When the proportion of hydrogen in the radiator increases, the high energy end of the response curve, which comes mainly from the radiator, is increased relative to the low energy part which is mainly from the etched layer. Figure 4 shows typical changes in the shape of the responses for thick polyethylene, CR-39 and Mylar radiators. The response does not increase as fast as the hydrogen content because an increased hydrogen content gives shorter proton ranges and so protons can only be collected from a smaller volume of radiator.

Figure 5 shows how the response varies for radiators of different thickness, given in mg/cm^2 by the numbers on the curves. At neutron energies for which the maximum range of recoil protons exceeds the radiator thickness, the response falls below that for a thick radiator and drops quite sharply. These calculated responses are for recoil protons only; because of the effects of heavy recoils and alpha particles the curves will flatten out

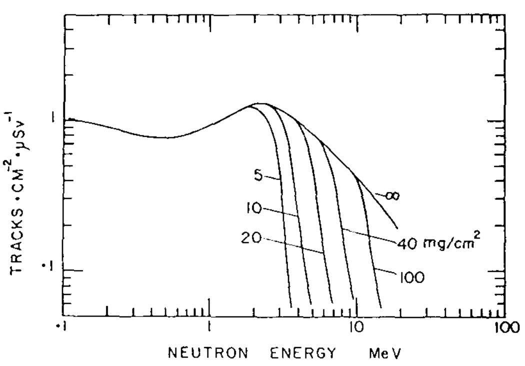

Fig. 5. Calculated Response of CR-39 Neutron Dosemeters for Various Polyethylene Radiator Thicknesses, Given in mg/cm² by the Numbers on the Curves.

at a few percent of the response for a thick radiator-as much as 25% in the case of 14 MeV neutrons.

There is another factor that complicates the response actually obtained with thin radiators — the fact that at high energies air acts as a reasonably efficient radiator[13]. Figure 6 shows how the response of CR-39 to 14 MeV neutrons varies with the thickness of a polyethylene radiator. Initially, the response drops as thickness increases, because the polyethelene absorbs more charged particles generated in the air than it contributed itself. The star shows the response from the etched layer alone, obtained with Au in front of the CR-39. At this energy, air provides over 50% as much response as a thick polyethylene radiator. For 2.8 MeV neutrons the relative contribution of air was only 3.5%. Most of this contribution is believed to come from (n,p) reactions in nitrogen.

2) Variation of Response with Angle of Incidence

Most of the following comments about angular response are not specific to electrochemically-etched dosemeters since, except perhaps at very low neutron energies, the angular response is determined by factors other than the etching technique. Figure 7 shows typical angular responses measured with a thick polyethylene radiator in front of CR-39. Similar responses for various

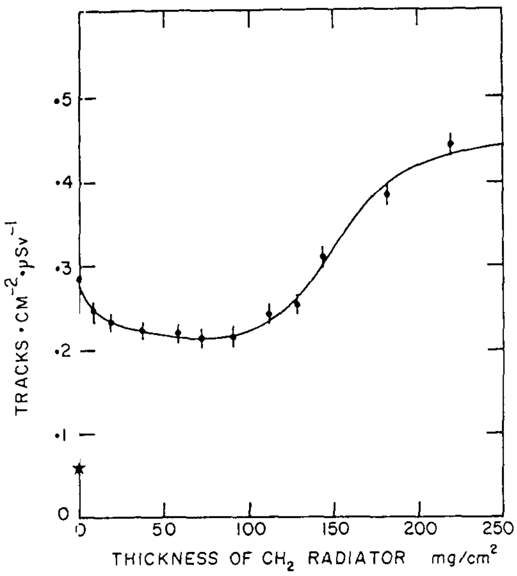

Fig. 6. Variation with Radiator (CH_2) Thickness of the Response of a CR-39 Dosemeter to 14.7 MeV neutrons.

neutron energies have been published by others[9,14-17]. The large variation with angle is the most serious defect of a CR-39 personal dosemeter.

The variation has two causes. The first is the existence of a maximum angle of incidence for detectable protons. Since the distribution of recoil protons directions is peaked in the neutron direction, normally-incident neutrons will generate more protons at angles within the critical angle than neutrons having other directions. This applies both to protons generated in the etched layer and those from the radiator. The second cause is the requirement for recoils produced in the radiator to escape into the CR-39. Consider a neutron beam incident on a thick radiator at angle θ. The number of neutrons striking 1 cm^2 of radiator varies as sin θ. If recoil protons were all in the neutron direction, the fact that the neutron path length through the radiator is increased would increase the number of protons generated along the neutron path but not the number of protons escaping

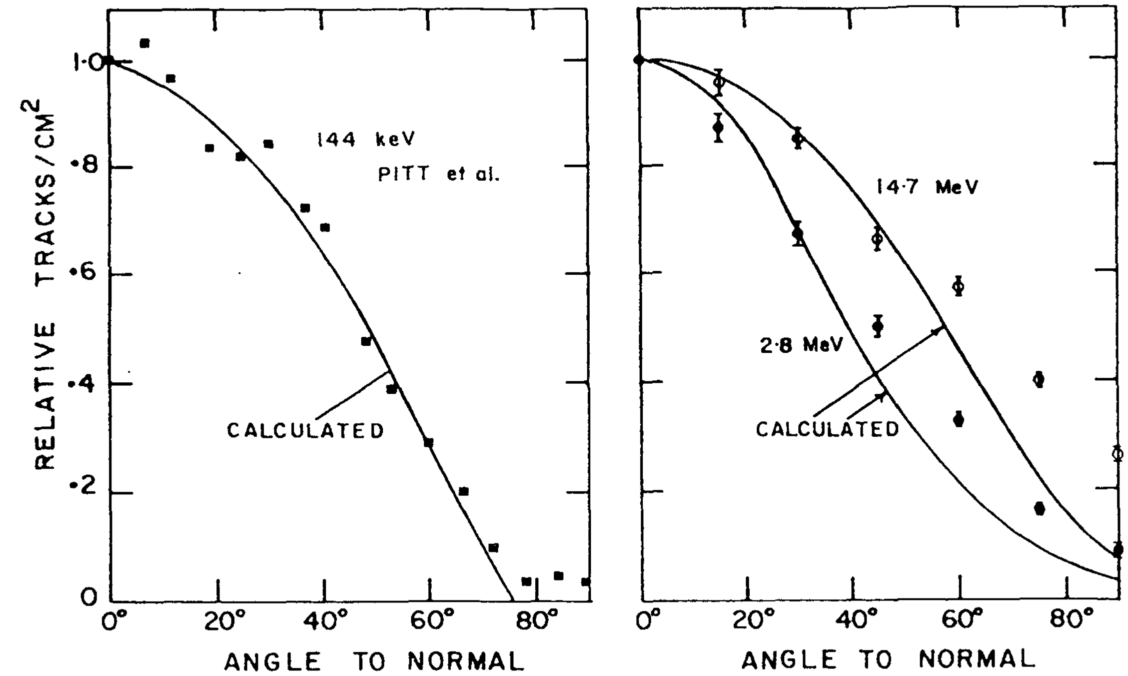

Fig. 7. Comparison of Measured (Points) Calculated (Curves) Angular Responses of CR-39 Dosemeters, for Neutrons of 144 keV (Data from Pitt et al, Reference 17), 2.8 MeV and 14.7 MeV.

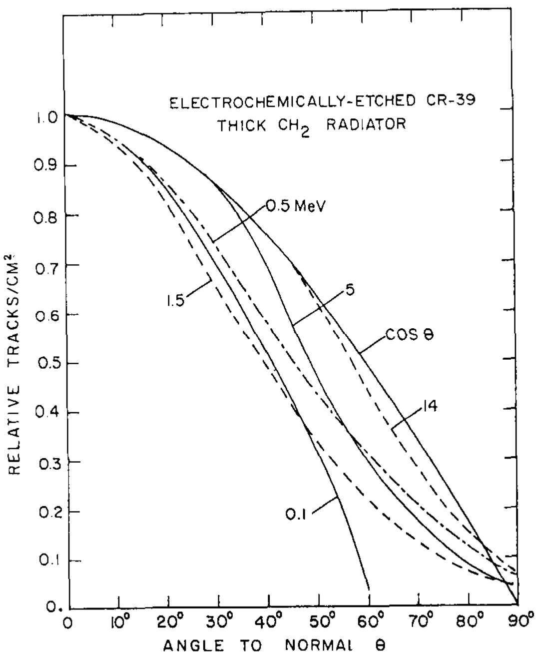

Fig. 8. Variation of the Calculated Relative Angular Response of a CR-39 Dosemeter with Neutron Energy, Given in MeV by the Numbers on the Curves. A Curve of cos θ is shown for Comparison.

into the CR-39, since only protons within a fixed distance of the boundary—namely the proton range—can escape. Then the number of escaping protons per neutron will be proportional to sin θ. Since tracks are only produced by protons with directions close to the normal, the angular distribution is more forward-peaked than sin θ.

The calculation of the angular distributions is quite straightforward and can be done on a desk-computer. Figure 8 shows calculated angular distributions for a number of neutron energies and a thick radiator, along with a curve of sin θ. It is difficult to see a steady progression of these curves from one energy to another because they are combinations of the effects of the radiator and of the etched layer and these two components have different angular distributions. Figure 7 shows a comparison for 144 keV, 2.8 MeV and 14.7 MeV neutrons. For 2.8 MeV the fit is not as good as we would have expected. At 14 MeV the measured value at large angles is expected to exceed the calculated value, because a significant fraction of the response comes from alpha particles and heavy recoils.

All the above angular responses are calculated or measured for a fixed neutron fluence. If we are trying to measure the individual dose equivalent at 1 cm depth, allowance must be made for the decrease of this quantity with increasing angle of incidence. Hence the CR-39 reading per unit individual dose euqivalent does not change significantly with angle, but the improvement is not very great over most of the energy range of interest.

The angular dependence can be changed slightly by using different radiator materials[18] since this varies the ratio of the contributions from the radiator and etched layer. Hankins et al. noted a simple way to improve the angular dependence[16]. When CR-39 is covered on both sides by polyethylene, the ratio of responses at 90° and 0° incidence is improved by more than 40% when tracks are measured on the "rear" surface of the CR-39, rather than on the front. This is easily understood.

The angular response can be changed much more by using a radiator thinner than the maximum range of recoil protons produced. This reduces the response at 0° and, as the angle of incidence increases, the radiator effectively gets thicker and the response is enhanced. While this might be a useful method if the neutrons were known to be monoenergetic, it is not practical for a dosemeter for general use. We cannot think of any practical method to improve the angular response as long as a planar detector is used. An alternative could be to combine the readings from three planar detectors mounted on the faces of a cube.

5. Conclusions

Calculation of the energy response of CR-39 dosemeters, using various factors that affect this response—the radiator, the etching conditions and the type of CR-39—shows many of the variations in response shapes that can be expected and helps one to understand measured effects. The calculated shapes fit measured shapes reasonably well but calculated values must sometimes be normalized to give the observed absolute track densities. Some measured data around 14 MeV do not fit calculated results but, on the basis of our own measurements at 14 MeV, we believe that these discrepant results are in error. Calculated angular responses have been less successful in fitting experimental data. Since these experiments are very simple and expected to be accurate, we are searching for possible errors in the theory.

References

1. Gibson, J. A. B. and Piesch, E. "Neutron monitoring for Radiological protection.", IAEA Technical Reports Series 252, pp. 63-71 (1985).
2. Hofert, M. and Piesch, E. "Neutron Dosimetry with Nuclear Emulsion", Radiat. Prot. Dosim., 10, pp. 189-195 (1985).
3. Hankins, D. W. "Factors Affecting the Design of Albedo Neutron Dosimeters Containing LiF Thermoluminescent Dosimeters". Los Alamos Scientific lab Rep., LA-4832 (1972).
4. Falk, R. B. "A Personnel Neutron Dosimeter Using Lithium Fluoride Thermoluminescent Dosimeters". Dow Chemical Co. Rep., RFP-1581 (1971).
5. ING, H. "Neutron Dosimetry Assessment". Prepared for Health Physics Society Summer School, Penn State University (1986).
6. Cartwright, B. G., Shirt, E. K. and Price, P. B. "A Nuclear Track Recording Polymer of Unique Sensitivity and Resolution". Nucl. Instrum. Meth., 153, pp 457-460 (1978).
7. Griffith, R. V. et al. "Monoenergetic Neutron Response of Selected Etch Plastics for Personnel Neutron Dosimetry". Radiat, Prot. Dosim., 1, pp. 61-71 (1978).
8. Tommasino, L. and Harrison, K. G. "Damage Track Detectors for Neutron Dosimetry: 1. Registration and Counting Method". Radiat. Prot. Dosim., 10, pp. 207-217 (1985).
9. Tommasino. L. et al. "Different Etching Processes of Damage Track Detectors for Personnel Neutron Dosimetry". Nucl. Tracks, 8. pp. 335-339 (1984).

10. Cross, W. G., Arneja, A. and Ing, H. "The response of electrochemically etched CR-39 to protons of 10 keV to 3 MeV". Nucl. Tracks, 12, pp. 649-652 (1986).
11. Benton, E. B., Oswald. R. A., Frank, A. L. and Wheeler, R. V. "Proton recoil neutron dosimeter for personnel monitoring". Health Phys., 40, pp. 802-809 (1981).
12. Harvey, J. R., Ramli, A. G. and Weeks, A. R. "Measurement of the etch characteristics of proton tracks in CR-39 and the implications for neutron dosimetry". Nucl. Tracks, 8, pp. 307-311 (1984).
13. Decossas, J. L., Vareille, J. C., Moliton, J. P. and Teyssier, J. L. "Theoretical study and calculation of the response of a fast neutron dosemeter based on track detection". Radiat. Prot. Dosim., 5, pp. 163-170 (1983).
14. Hadlock, D. E., Parkhurst, M. A. and Haggard, D. L. "High energy neutrons interact with air! Does CR-39 respond?" (Abstract) Health Phys., 49, p. 143 (1985).
15. Bartlett, D. T. and Steele, J. D. "The energy and angle dependence of response of the NRPB CR-39 fast neutron dosemeter". In Proc. Fifth Symposium on Neutron Dosimetry, Vol. 1 (H. Scharube, G. Berger and J. Booz, eds.), CEC Report EUR 9726 EN, Luxembourg, pp. 511-518 (1985).
16. Piesch, E. and Urban, M. "Dosimetric properties of different CR-39 plastics used as neutron recoil track etch detector". Nucl. Tracks, 12, pp. 661-664 (1986).
17. Hankins, D. E. and Westermark, J. "Neutron spectra evaluation obtained from the track size distribution on electrochemically-etched CR-39 foils". Radia. Prot. Dosim., to be published.
18. Piesch, E., Jasiac, J. and Urban, M. "Markrofol and CR-39 recoil track etch detectors as a supplement of a universal albedo neutron dosimeter". Nucl. Tracks, 8, pp. 323 (1984).