CONF-721203--2
RECEIVED BY TIC OCT 31 1972

PROGRESS IN SOLID-STATE FAST NEUTRON PERSONNEL DOSIMETRY*

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ABSTRACT

The poor stability of latent recoil proton tracks in photographic emulsions has frequently been underestimated due to unrealistic testing conditions. Thin polycarbonate foils to be evaluated by automatic spark-counting exhibit no fading for at least three months at 32°C and 95 % humidity. As one of several alternatives to the NTA film, optimized combinations containing ^{237}Np or ^{232}Th and 10 μm polycarbonate foils are described (^{237}Np covers a wide dose range of $\sim 10^{-3}$ to ~ 10 rad, and neutron energies ~ 0.6 MeV). The directional response is given for different neutron energies and detector-phantom distances. Another approach consists of the amplification of recoil nuclei etch pits to facilitate automatic counting. The inherently low fast neutron response of most solid-state detectors can be increased by intimate contact with a hydrogenous material. TSEE detectors based on BeO, for example, which are covered with a hydrogenous radiator, have been used successfully in recent tests of criticality monitoring systems at ORNL. The thin sensitive layer of exoelectron detectors makes this approach especially attractive for intermediate neutron energies.

*Research sponsored by the U.S. Atomic Energy Commission under contract with the Union Carbide Corporation.

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Introduction

Much of the data which have been accumulated during the last thirty years of large-scale personnel neutron dosimetry are highly questionable for reasons of insufficient accuracy of the dosimeters and oversimplified interpretation of the results, and some guidelines on the desirable accuracy of personnel dosimeters are (as has already been pointed out in some detail in the paper presented by J.A. Auxier) badly needed. In Fig. 1, a proposal for accuracy requirements for personnel dose measurements as a function of dose is given as a possible basis for further discussion [1]. (It should be noted that the given errors, which may appear to be quite large, are not those errors one may obtain in tests under idealized laboratory conditions with frontal free-air exposures to monoenergetic neutrons, etc., but total errors including directional and energy dependence of the dosimeter response when placed on the body, fading during extended monitoring periods, and all other potential sources of inaccuracy).

Thermal neutrons have the advantage of being easily measurable, but they are *per se* only of very limited interest in personnel neutron dosimetry - there are very few, if any, cases in which a substantial part of a person's external radiation exposure has been due to thermal neutrons. Thermal neutron detectors have, nevertheless, been used extensively in some nuclear installations to indicate the presence of intermediate and/or fast neutrons based on the albedo principle. Frequently, the inherent inaccuracy of albedo dosimeters (due mostly to their strong energy dependence and the effect of distance from the body) has been underestimated, but encouraging improvements have been made recently. A device was developed in Savannah River [2] which also incorporates some features of the multisphere technique and was found to be quite accurate for a moderated fission spectrum up to about 0.6 MeV. Although such systems help to solve the monitoring problems in the intermediate energy range, for example for plutonium workers, there remains an urgent need for a reliable, stable and sensitive fast neutron personnel dosimeter.

Photographic Track Emulsions

There has been mounting evidence in the past years that the use of nuclear track emulsions in fast neutron dosimetry should be discontinued even if no alternative methods are available, because their extremely limited benefits do not justify the substantial manpower cost of track film evaluation. There are various more or less well-known contributing factors, which make the occasions where reasonably accurate results can be obtained with this method very rare indeed. For example, many of the earlier calculations on the low-energy response limit of track films have been based on the assumption that short recoil proton tracks consisting of three or four grains can be counted; it has, however, been shown [3] that there is no relationship whatsoever between the count of such "tracks" and the neutron dose.

More importantly, the extent of latent track fading which takes place in all but extremely dry and cool desert climates is still not fully realized by many operators of film badge services. Partly this is due to sheer ignorance, and partly

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Fig. 1 Suggested maximum tolerable total error in the personnel dosimeter indication, and the consecutive evaluation of the actual average or critical organ(s) dose.

due to unrealistic conditions under which fading tests have frequently been carried out. One of the facts which have been overlooked in many such tests is that the film, if worn by a person, has usually been in equilibrium with humid air for at least days, often weeks before the neutron exposure occurred. As can be seen in Fig. 2, it takes some time before the humidity penetrates the wrapping of a dry film, and rapid fading is observed if the emulsion is permitted to equilibrate. The comfortable relative humidity is around 50 to 75 %, and in large parts of the world the humidity as well as the ambient temperature is even higher during at least part of the year. Under such conditions, 50 % fading occurs within less than two days, and no tracks would be left at the end of a monitoring period [4] (Fig. 3).

It is widely believed that sealing of the dry film into some polymer foil remedies this problem sufficiently. One should, however, be aware that

- a. the permeability of polymers to water vapor varies widely and strongly depends on such factors as their density, crystallinity, orientation and cross-linking of the molecules, and additives; as shown in Table I, ethyl cellulose is, for example, by over a factor of 10^4 more permeable than certain types of Teflon [5];
- b. even the most impermeable film has a measurable permeability, and the rate of water vapor transmission depends on the vapor pressure gradient across the film, with the driving force being largest with a completely dry film and 100 % relative humidity outside; and
- c. the relatively best results are to be expected with a metal foil-plastic-compound "vapor barrier" which is very carefully sealed, leaving a wide seam (as an additional measure, a dessicant may be included into the package).

Considering these, and some other limitations of the track film with regard to the interference of gamma fogging, its limited dynamic range, the difficulties in automating evaluation, etc., it is obvious that its use should be restricted to short-term experiments around a few high-energy neutron sources with very low gamma background. Unfortunately, progress in the development of replacements for the track film has not been as rapid in recent years as one would have hoped. However, some results are encouraging.

Track Etching

In track etching, one of the two possible approaches; namely the determination of fast neutron induced recoil carbon, oxygen, and nitrogen nuclei; is still hampered by the fact that the resulting tracks or etch pits are small and short, thus making it impossible to use the elegant method of spark counting for the determination of low track densities in large detector areas [6]. External "radiators" providing alpha particles by threshold (n,α) reactions contribute only little to the inherent sensitivity of the foil if track counting is done visually after conventional etching [7] (one notable exception is the exposure of foils to fast neutrons in a helium atmosphere, which produced a twelve-fold increase in tracks). Even if thin, alpha-sensitive cellulose nitrate foils and spark-counting techniques are employed for automatic evaluation [6] this approach is presently of little practical interest due to the small cross-section of most fast neutron induced (n,α) reactions.

Fig. 2 Fading of latent recoil proton tracks from a Po/Be source in the standard Kodak NTA film at 75 and 90 % relative humidity; with the film originally dry and then stored at the indicated humidity; and with the film in equilibrium with its environmental humidity (after Amadesi *et al.*, Min. Nucl. 4, No 11, 1960, and 5,323, 1961; and Becker, Atomkernenergie 8, 74, 1963).

Fig. 3 Fading of latent radiation effect in Kodak NTA film during storage in a protected space outdoors and in the laboratory in a semi-tropical climate (during May/June 1971 in Taiwan).

Table I. Permeability of Polymers to Water Vapor

Polymer	P^x
Ethyl cellulose	12,000
Cellulose acetate	5,500
Polycarbonate	1,400
Polystyrene	1,200
Polyvinyl chloride	156
Polypropylene	51
Polyethylene (high-pressure)	12
Polyvinylidene chloride	0.5
Polytrifluorochloro- ethylene (Teflon)	0.29

$$x) P = \frac{(\text{amount of permeant}) (\text{film thickness})}{(\text{area}) (\text{time}) (\text{pressure-drop across film})} \times 10^{10} \text{ (at 20-25°C)}$$

It has been shown recently [8] that the size of the damaged area in the environment of a track or etch pit can be increased with an electrochemical etching procedure [9]. Unfortunately, the resulting spots from recoil nuclei (Fig. 4) are much less well defined than those of fission fragments. There are presently only two choices for the evaluation of foils for recoil and (n, α) reaction products: One is the slow and tedious visual counting, the other the cumbersome and complicated automatic evaluation with electro-optical systems and a computer. It should, however, be noted that the recoil nuclei etch pits in the plastic backing of the photographic NTA film can be used, after removal of the emulsion, as a good fast neutron accident dosimeter. Other etchable polymers worn by the exposed person including the frames of eye-glasses, plastic badges, etc., may also be used for this purpose.

The spark-counter for the automatic counting of fission fragment tracks in thin polycarbonate foils has reached a relatively high level of reliability. Track densities up to 5000 tracks/cm² can be counted with less than 10 % counting loss, and reproducibilities of better than 1 % have been reported [10]. Of the various fissionable materials, ²³²Th is of limited interest because of its penetrating gamma radiation and the high detection threshold of about 1.6 MeV (Fig. 5); ²³⁸U has an unacceptably high spontaneous fission rate, and the use of ²³⁵U is subject to complicating restrictions in many countries, as well as having a high thermal neutron response.

Neptunium-237, having a detection threshold of ~ 0.6 MeV and a large fission cross section, is the most attractive fissionable material, but care has to be taken to minimize its potential radiological hazards in personnel dosimeters. Neither electroplating nor vacuum deposition has resulted in well-sticking, uniform layers of the desirable thickness (2-5 mg/cm²). The best results so far have been obtained by "burning-in" several layers of evaporated organic solutions of neptunium nitrate onto a metal disk at high temperatures, resulting in a usable good neptunium oxide layer.

The ²³⁷Np layer is additionally protected from potential rub-off of alpha activity by sealing into a 2 μ m polycarbonate foil. This results in a sensitivity drop of about 20 % (Fig. 6). However, the sensitivity range of a detector containing about 8 mg of ²³⁷Np still permits measurements in the dose range from about 1 millirad (six sparks, no background sparks in the unexposed foil) to about 10 rads of HPRR fission spectrum neutrons (higher doses can be evaluated by microscopic track counting). Reduction in the thickness of the ²³⁷Np layer, or replacement of the neptunium by thorium leads naturally to a decrease in sensitivity.

The fading stability of latent fission fragment tracks in polycarbonate prior to etching and spark-counting is satisfactory. Even at very high humidity, no fading has been observed during three months at 30°C (Fig. 7), and only 25 % fading occurred during three weeks at 60°C [11]. The fading becomes more pronounced [12] if the storage temperature exceeds 100°C.

There is, of course, a strong effect of the human body on the reading of any dosimeter if worn on the front of the trunk and a monodirectional exposure occurs from the side or back. As an example, the situation is described for 14 MeV

Fig. 4 Microphotograph of fission fragment (left) and recoil nuclei (right) etch pits in cellulose acetobutyrate (backing of Kodak NTA films) enlarged by electrochemical etching.

Fig. 5 Fission cross sections of ^{237}Np , ^{238}U , and ^{232}Th as a function of neutron energy.

Fig. 6 Fission fragment spark counts of etched polycarbonate foils as a function of neutron dose from a fast reactor (HPRR) with different radiators and plastic covers.

Fig. 7. Spark count in fission-fragment irradiated polycarbonate foils as a function of storage time at elevated temperatures and high humidity between exposure and etching.

neutrons in Fig. 8. The directional effect naturally becomes more pronounced with decreasing neutron energy. Varying the distance between dosimeter and body between 0 and 2 cm has, however, only little influence on the results. A special variation of the track etching personnel dosimeter is a ring dosimeter (Fig. 9) to be used for monitoring the fast neutron hand exposure of persons working with plutonium or californium in glove boxes, doing ^{252}Cf needle implantation work in neutron cancer therapy, etc. (see also ORNL-4795, p. 40).

Exoelectron Dosimetry

Unlike track etching, the application of thermally stimulated exoelectron emission (TSEE) in fast neutron dosimetry is fairly new [13]. A problem which had to be solved before any practical application could be considered was the relatively poor stability and reproducibility of early TSEE systems. A new counter has been developed at ORNL for ceramic BeO detectors which not only simplifies the readout of large numbers of detectors with a special loading procedure, but also helps to avoid many of the earlier problems by the application of a "pushing" electrical potential which injects the low-energy exoelectrons into the counting volume during readout [14]. It was thus possible to evaluate such detectors not only with a high sensitivity of more than 10^3 counts for one mrad per cm^2 of emitting surface area, but - more importantly - to reduce the standard deviation of multiple readings to 2-3 %. As an example, the effect of the accelerating potential is illustrated in Fig. 10.

Of course, BeO has a low inherent sensitivity to fast neutrons. However, pairs of BeO disks - one covered with polyethylene or another hydrogenous material, and the other covered with graphite or Teflon as low-Z non-hydrogenous substance - exhibit a pronounced difference in response in mixed radiation fields due to the effect of recoil protons from the polyethylene (Fig. 11). Because of the extremely thin sensitive layer of TSEE dosimeters, they permit the registration of rather low-energy recoil protons. The recoil proton "radiator" can be composed of either gaseous (e.g., an organic gas), liquid, or solid hydrogenous materials. There is a slight increase in the fast neutron sensitivity of this system from ~18 % at 100 keV, to ~28 % of the gamma radiation response at 16 MeV [13].

In the last Intercomparison Study of accident monitoring systems at ORNL, there was good agreement between dose measurements with TSEE detectors and the values obtained by other systems [15] (Table II). It should, however, be noted that, as the neutron to gamma ratio decreases, the accuracy of the neutron measurement also decreases.

With the recent progress in the development of TLD detectors such as $\text{CaSO}_4:\text{Tm}$, $\text{CaSO}_4:\text{Dy}$, and $\text{MgSiO}_4:\text{Tb}$, which combine a sensitivity to ionizing radiation 20 to 50 times higher than LiF:Mg,Ti with an excellent fading stability, the old idea of using fine-grained TLD materials in intimate mixture with organic recoil proton "radiators" also deserves some new attention. The two constituents of such a system can either be separated before readout - a mixture of the TLD phosphor with sugar is an example for this approach [16] - or they can remain together if the hydrogenous material is sufficiently heat-resistant. This latter possibility is presently being studied at ORNL.

ORNL-DWG 71-6139

Fig. 8 Directional response of dosimeters based on ^{232}Th and ^{237}Np /Kimfol combinations, if exposed on the surface of a water-filled thorax phantom to monodirectional 14 MeV neutrons.

Fig. 9 Open and closed track-etching finger-ring dosimeter for hand monitoring (this version containing a ^{232}Th foil ~ 10 mm in diameter, in contact with a $10\text{ }\mu\text{m}$ polycarbonate foil glued with rubber cement to a nylon retainer ring to simplify marking and handling during etching and sparking).

ORNL-DWG 72-11267

100 pA ^{137}Cs γ

1/2" BeO 995

HEAT SENSITIZED $\sim 1400^\circ\text{C}$

FOR 264 HOURS AND STABILIZED

IN AIR AT 80% RELATIVE HUMIDITY

MEAN RESPONSE $\bar{I}$ (NET COUNT 200-400°C)

$= 104,200$

STANDARD DEVIATION σ

$= 3.0\%$

-67.5 VOLTS

0 VOLTS

TSEE RATE (cps)

$\bar{I} = 32,600$

$\sigma = 4.7\%$

100 200 300 400
SAMPLE BASE TEMPERATURE ($^\circ\text{C}$)

Fig. 10 Recordings of the TSEE peak of ceramic BeO discs (Brush Thermalox 995, 12.5 mm diameter) before (lower curves), and after (upper curves) applying of a -67.5 V "ejection" potential to the back of the disc during linear heating.

Fig. 11 Principle of TSEE fast neutron dosimetry: One BeO dosimeter is covered with a non-hydrogenous material, another with a hydrogenous material; the difference in the response in a mixed radiation field indicates the neutron-induced recoil proton dose (the absolute sensitivity indicated in this graph has since been increased).

Table II. Results of TSEE Dosimeters in Comparison with the Average of other Detectors (in brackets) during 1972 ORNL Nuclear Accident Dosimetry Intercomparison Study

HPRR Burst No.	shielding (cm)	dose (rads)	
		neutron	gamma
1	-	250 (256)	36 (35)
2	13 steel	108 (110)	13 (13)
3	12 Lucite	42 (45)	32 (36)
4	-	263 (272)	40 (41)

Acknowledgments. This paper, which reviews some recent work of the ORNL Solid-State Dosimetry Research Group, contains some unpublished (or to be published) data from several other members of the group, in particular Dr. R.B. Gammage, J.S. Cheka, M. Lee, and M. Sohrabi. T.G. Clark participated in the design and testing of the ring dosimeter.

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Figure Captions

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