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A neutron dosimeter based on ^{237}Np fission

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A NEUTRON DOSIMETER BASED ON ^{237}Np FISSION

K.G. Harrison

ABSTRACT

A new type of personal dosimeter for neutrons based on the detection of fission-fragments from ^{237}Np by a thin-film plastic track detector is proposed as a potential replacement for the nuclear track emulsion. Fission-fragment tracks in the plastic film are etched into holes by caustic alkali and can be counted very rapidly using a spark counter. The new dosimeter contains a sealed source of 4 mg of neptunium, and has a good dose-equivalent response and useful sensitivity down to less than 30 mrem. The localized surface skin-dose to the wearer from gamma-radiations emitted by the source is reduced to about 11 mrem/40 h by an integral radiation shield and the gonad dose-rate for a dosimeter worn at waist level is about 2 mrem per working year. The dosimeter presents administrative problems and can only be recommended for use in radiation areas having high security.

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Fig.

1. The theoretical dose-equivalent sensitivity of ^{237}Np to neutrons (free-air)
2. The theoretical dose-equivalent sensitivity of ^{237}Np to gamma radiation (free-air)
3. Exploded view of the proposed dosimeter.

1. Introduction

The existing personal dosimeter for fast neutrons is the nuclear track emulsion. This has a number of shortcomings, of which the variable fading of undeveloped tracks and the slow readout with an optical microscope are probably the most important. Practical alternative dosimeters are based on either the thermal-neutron albedo effect with neutron-sensitive and neutron-insensitive TLD's (Harvey et al., 1973), or (n,fission) reactions with fission-fragments detected in plastic track recorders such as polycarbonate (Becker, 1972). However, because of the shape of the fluence to dose-equivalent conversion function for neutrons (ICRP, 1973), the dose-equivalent sensitivity of all simple thermal-neutron albedo systems drops sharply above about 10 keV (Harvey, 1968).

Fission dosimeters have adequate dose-equivalent sensitivity for fast neutrons, but generally have high thresholds (~ 1 MeV), or, alternatively, have overwhelming thermal and intermediate neutron responses. A fission dosimeter using ^{237}Np is unique in having a response approaching the current dose-equivalent requirement (ICRP, 1973) over a very wide range of neutron energies (Harrison et al., 1977), but ^{237}Np has the disadvantage of fairly high specific activity compared with thorium or natural uranium. A personal dosimeter using this material must therefore have sound containment and efficient radiation shielding for the wearer.

The fission dosimeter is simple in principle, and is well described in a review by Becker (1972) and more recently by Cross and Ing (1975). A thin plastic film, typically $10\text{ }\mu\text{m}$ polycarbonate, is held close to the fissile source within the dosimeter. Incident neutrons produce fissions, and the resulting fission-fragments pass through the plastic film creating stable but invisible damage tracks. The film is then removed from the dosimeter and etched in concentrated caustic alkali. The etching-rate of the damage tracks is much higher than that of the bulk material and each track is etched into a hole several microns in diameter. These holes are then counted optically or using a spark counter (Cross and Tommasino, 1970; Harrison et al., in preparation).

In the following sections, the choice of ^{237}Np as the fission source is justified and its response is evaluated both theoretically and experimentally. Possible interfering effects and practical problems are then discussed, and a suggested design for a dosimeter using ^{237}Np is outlined.

2. Theory

2.1 The choice of fissile material

Unfortunately, all practical materials which undergo fission following neutron capture are also radioactive alpha-emitters. The acceptability of any isotope therefore depends on its spontaneous-fission rate, alpha activity, gamma-ray activity and toxicity as well as its expense, availability, purity and the correspondence between its (n,f) cross-section and the required dose-equivalent response. All isotopes with large thermal-neutron fission cross-sections also have large cross-sections at epithermal and intermediate neutron energies, and therefore over-respond to all these energy ranges (relative to fast neutrons) by several orders of magnitude. Although the thermal and epithermal responses can be eliminated using suitable neutron filters it is not possible to reduce adequately the response to intermediate-energy neutrons.

Of the remaining isotopes, only ^{232}Th and ^{237}Np have acceptably low spontaneous-fission rates for long term dosimetry ($2\text{ fissions mg}^{-1}\text{a}^{-1}$ for ^{237}Np and $< 10^{-2}\text{ fissions mg}^{-1}\text{a}^{-1}$ for ^{232}Th). ^{232}Th has a much lower specific activity than ^{237}Np ($10^{-4}\text{ }\mu\text{Ci mg}^{-1}$ compared with $0.7\text{ }\mu\text{Ci mg}^{-1}$), but it has a much higher fast neutron threshold (1.3 MeV compared with 600 keV) and has negligible sensitivity in the energy region below 30 keV, whereas ^{237}Np fortuitously has a near dose-equivalent response in this range (see Fig. 1). It seems therefore that although ^{232}Th would be easier to use than ^{237}Np , it is by comparison extremely limited in its applicability.

2.2 Cross-sections for ^{237}Np

2.2.1 (n,f) cross-sections

The cross-sections for the (n,f) reaction in ^{237}Np have been extensively studied over the range of neutron energies from about 100 keV to 14 MeV and are well known (Schett et al., 1974; Bak and Lorenz, 1971; Magurno, 1975). Below 100 keV the cross-sections are small (≈ 20 mbarns) and there are relatively few measurements and compilations (Garber and Kinsey, 1976). The situation is complicated by the fine resonance structure in the cross-sections in this low-energy region, and care must be exercised when integrating them with neutron spectra to obtain theoretical responses.

Useful published data which include sub-threshold cross-sections are confined to three main sources. These are the experimental measurements of Jiacoletti and Brown (1971) and evaluations by Simons and McElroy (1970) and Delafield et al. (1973b). The last two have differences of up to 60% in the range from 100 eV to 50 keV, but are in better agreement with each other than with Jiacoletti and Brown's measurements, which are higher by up to a factor of about two over most of the sub-threshold energy range. The cross-sections tabulated by Delafield et al. (1973b), which are a graphical interpolation of three sources (Stehn et al., 1965; Davey, 1968; Gavrilov et al., 1970) are found to be in best agreement with the present experimental measurements (Section 3; Table I). These cross-sections are plotted in Fig. 1 in units of fissions rem^{-1} of neutrons for 4 mg of neptunium, with fluence to dose-equivalent conversion factors taken from the ICRP recommendations (ICRP, 1973).

2.2.2 (γ ,f) cross-sections

The photofission reaction for ^{237}Np does not seem to have been widely studied. The available data (Katz et al., 1958; Huizenga et al., 1962; Veyssière et al., 1973) have been studied by Patrick (1975) and his assessment of the likely photofission response is plotted in Fig. 2, in units of gamma-ray dose-equivalent.

It can be seen that above 5 MeV the gamma-ray response is significant. In areas around reactors where high-energy gamma radiation occurs (especially from ^{16}N decay) it is possible that the dosimeter may record gamma-ray dose rather than neutron dose. According to Blizard and Abbot (1962), fission gamma-ray spectra from bare sources, including prompt, delayed and capture gamma-rays, produce a dose component from photons above 5 MeV which is about three orders of magnitude less than the fast neutron dose-equivalent, so that for bare fission sources the gamma-ray response is unlikely to be a problem.

2.3 The albedo-neutron response of the ^{237}Np dosimeter

Since ^{237}Np has a finite (n,f) cross-section at thermal and intermediate neutron energies, it is expected to show a response to the backscattered (albedo) neutrons, when worn close to the body. The magnitude of this response relative to the response to incident neutrons will depend on the relative cross-sections and the backscatter spectrum and intensity, and may form a considerable fraction of the total response. The well-known thermal-neutron albedo based on the backscattered flux of thermal neutrons (see, for example, Smith, 1966; Harvey, 1968), used with the thermal-neutron fission cross-section, will probably be inappropriate, because the backscatter-spectrum is not entirely thermalized (Alsmiller and Barish, 1974), and the fission cross-section for neptunium is smaller at thermal-neutron energies than at intermediate energies.

The overall response of a pure ^{237}Np dosimeter (worn on the body) to plane-parallel normally-incident monoenergetic neutrons of energy E , with no neutron filters and no flux depression, must therefore be derived from the full equation:-

$$F(E) = \Phi(E).n.\sigma(E) + \beta(E).\Phi(E).n \int_0^E \sigma(E')\phi_E(E')dE'$$

where $F(E)$ = the number of fissions
 $\Phi(E)$ = the monoenergetic neutron fluence at energy E
 n = Number of ^{237}Np atoms ($4 \text{ mg} \equiv 1.02 \times 10^{19}$ atoms)
 $\sigma(E)$ = (n,f) cross-section for ^{237}Np
 $\beta(E)$ = total albedo backscattered fraction for incident neutron energy E
 $\phi_E(E')$ = backscattered albedo spectrum for incident neutrons energy E ,
normalized such that

$$\int_0^E \phi_E(E')dE' = 1$$

The first term is the direct (free-air) response and the second term is the albedo (backscatter) response. Unfortunately, neither $\phi_E(E')$ nor $\beta(E)$ are well known, although some calculations of $\beta(E).\phi_E(E')$ up to $E' = 17 \text{ eV}$ (albedo-neutron energy) have been made (Alsmiller and Barish, 1974), and other calculations are in progress (Gibson, 1976).

2.4 Angular anisotropy of fission-fragment emission

The angular distribution of fission-fragments from neutron-induced fission is not isotropic. For ^{237}Np , measurements have been reported in the neutron energy range from 100 keV to 7 MeV (Androsenko et al., 1969) which show a marked deviation from isotropy with increasing neutron energy, up to about 15% at 1 MeV and 25% at 6 MeV. Below 100 keV, no measurements are available, but for this sub-threshold region there appears to be no mechanism which would theoretically give directional anisotropy.

The nature of the fission detector in the proposed dosimeter, in which flux-depression is negligible for all energies of neutrons, suggests that fission-fragment anisotropy of up to 20-30% may lead to a dependence of the spark-counter efficiency (spark counts/fission) on the neutron energy, for normally incident neutrons, as well as an angular dependence of the dosimeter for high-energy neutrons.

3. Measurements of the Neutron Response of the Dosimeter

Measurements have been made with ten sources of neutrons covering a wide range of neutron energies between thermal and 10 MeV. Experimental dosimeters (Harrison et al., in preparation), each containing a thin bare source of 4 mg of ^{237}Np in contact with a 10 μm polycarbonate fission-track detector, were used for the exposures. These were generally undertaken with dosimeters in free-air and also on a "bomab" phantom (Delafield et al., 1973b, Fig. 6). In all the irradiations the dosimeters were oriented such that the polycarbonate was behind the neptunium. Some angular-dependence measurements were also made in free-air and on-phantom for the ^{252}Cf and Pu-Be sources. Details of the irradiation arrangements, fluence measurements and neutron spectra for the individual sources are given in each of the following experimental sections (3.1-3.5).

The track detectors were etched in 7.1 N KOH solution at 60°C for 60 mins, and the number of fission tracks counted using a spark counter. The details of the processing and counting procedures are fully described elsewhere (Harrison et al., in preparation).

3.1 Thermal neutrons

Thermal-neutron detectors are frequently calibrated by subtracting the response of the detector under cadmium from the response of the bare detector, following equal exposures in a well-thermalized neutron flux. For ^{237}Np this procedure is inaccurate because of photofission of neptunium by the high-energy gamma radiation from the $\text{Cd}(n,\gamma)$ capture reaction (Cross and Ing, 1975).

To avoid this problem, a moulded epoxy box with a wall-loading of 80 mg cm^{-2} of ^{10}B in the form of natural boron carbide was used in place of a cadmium shield. This loading produces a neutron cutoff below about 0.4 eV, where its transmission is 0.01. However, the wall-thickness of the box is sufficient to perturb the incident fast-neutron spectrum, so the "bare" irradiation was done in a plain epoxy box having the same dimensions as the boron-loaded box. The irradiations were undertaken at the thermal column of the GLEEP reactor.

Preliminary measurements with pairs of LiF thermoluminescent chips (Harshaw TLD-600 and TLD-700) indicated that the thermal-neutron flux inside the plain box was $(10 \pm 5)\%$ higher than the incident thermal flux, and this correction was applied to the subsequent calibrations. The incident fluences were measured using gold foil packs from UKAEA criticality lockets (Delafield et al., 1973a), positioned in free-air each side of the boxes.

For the thermal column of GLEEP the response of the dosimeter in the boron-loaded box was only 8% of that in the plain box. The on-phantom response could not be measured experimentally because of the low neutron flux at distances from the reactor where repeated cross-scattering would be negligible, but in common with all thin thermal-neutron detectors, it should be 1.8 times higher than the free-air response (see, for example, Harvey, 1968).

3.2 Sb-Be neutrons (0.5 keV and 23 keV)

Experiments were undertaken in the large shielded neutron calibration cell at the CEGB Nuclear Laboratories, Berkeley, UK (BNL), in which dosimeters were irradiated for many hours in free-air and on-phantom using a bare Sb-Be source ($1 \times 10^7\text{ n s}^{-1}$ emission) and a moderated Sb-Be source ($2 \times 10^6\text{ n s}^{-1}$ emission). These experiments are fully described elsewhere (Harrison et al., 1977). The emission rates of the sources were determined using a de Pangher long counter, which had been calibrated previously with these types of sources against the BNL manganese bath system (Harvey et al., 1976).

According to Ryves and Robertson (1971) a bare Sb-Be source emits 95.6% of neutrons at 23 keV and 4.4% at 375 keV. From these figures it is calculated (using cross-sections from Delafield et al. (1973b)) that the contribution of the high-energy neutrons to the total number of fissions is theoretically 27% for the free-air measurement and, using a phantom/free-air ratio of 1.4 for 375 keV neutrons (Section 3.3), is only 10% for the on-phantom result. The response due to the 23 keV component alone can therefore be derived.

The neutron spectrum of the moderated source has been evaluated theoretically using Monte Carlo methods (Harvey and Bending, 1976). Neutrons are emitted with a continuous range of energies from about 5 eV to 375 keV, with a broad peak at 500 eV in a lethargy spectrum. It has not yet been possible to verify this calculation experimentally.

3.3 Accelerator experiments using $(p + ^7\text{Li})$ and $(p + ^3\text{T})$ reactions (100 keV-1.72 MeV neutrons)

Neutrons were produced by the $^7\text{Li}(p,n)^7\text{Be}$ and $^3\text{T}(p,n)^3\text{He}$ reactions using protons from the IBIS 3 MeV accelerator at Harwell. A low-scatter facility was used so that shadow-cone corrections for the dosimeters at all the irradiation energies were less than 5%. The exposure at 100 keV was made at 30° to the direction defined by the ion-beam, but all the other exposures were at 0° .

Because of the low intensities of the sources, free-air irradiations were generally made at 20 cm and on-phantom irradiations at 40 cm from the accelerator target. One on-phantom measurement at 80 cm with 0.55 MeV neutrons agreed within experimental error with the 40 cm result at the same energy, and it is thought that the diverging neutron beam introduces only a small deviation in the albedo-neutron response from the ideal uniform parallel neutron beam. Nevertheless, the 100 keV measurement was made at 90 cm because of the particular importance of the albedo effect at this energy.

Neutron fluences were measured using a de Pangher long counter which had been calibrated at the National Physical Laboratory at five energies in the range 140 keV-5.21 MeV and shown to have the same response (within 2%) as their standard de Pangher long counter. This counter was used to calibrate the beam integrator and a second long counter (at 40° and 3 metres from the target), which were used as monitors during the irradiations.

The neutron energies were derived by operating the accelerator in its pulsed mode, when conventional time-of-flight spectra were obtained using a ^6Li -loaded glass scintillator for the neutron detector. The spectra were converted to neutron energy spectra using a calculated detection efficiency for the scintillator.

3.4 ^{252}Cf neutrons ($\bar{E} = 2.13 \text{ MeV}$)

A ^{252}Cf neutron source having an emission rate of about $6 \times 10^6 \text{ n s}^{-1}$ (calibrated in a recent comparison with an NPL standard source, using a long counter) was used to measure the response of the dosimeter in free-air and on-phantom. Exposures were made in low-scattering conditions at 15 cm from the source.

Additional exposures were made on-phantom at several distances between 20 and 70 cms to investigate the variation of the albedo response with the divergence of the neutron beam. The relative variation of the response of the dosimeter at three angles of incidence was also measured in both free-air and on-phantom exposures. These measurements were only made with the dosimeters at 15 cm from the source, because of its low intensity. Responses at 90° were not measured as these would be strongly perturbed by the stainless-steel casing of the dosimeter.

The neutron spectrum of the source was assumed to be Maxwellian, with a mean energy of 2.13 MeV (Werle and Bluhm, 1972).

3.5 Pu-Be neutrons ($\bar{E} = 4.04 \text{ MeV}$)

A Pu-Be source with an emission rate of $3.73 \times 10^6 \text{ n s}^{-1}$ (recently calibrated against an NPL standard Am-Be source using a long counter) was used to measure the response of the dosimeter to the 1-10 MeV neutrons of the Pu-Be spectrum. Exposures were made at 15 cm with the dosimeters both in free-air and on-phantom, in low-scattering conditions. Angular variations in the responses in free-air and on-phantom were measured as for the ^{252}Cf source, and an additional measurement was made of the response to Pu-Be neutrons passing through the phantom.

Because of uncertainty about the neutron spectrum of this source, values of 1.48 barns for the mean fission cross-section and $2.77 \times 10^7 \text{ n cm}^{-2} \text{ rem}^{-1}$ for the effective conversion factor, given by Burger et al. (1971) for a similar source, were used.

3.6 Results

The results are summarised in Table I with the additional angular and distance dependence measurements for ^{252}Cf and Pu-Be sources in Tables II and III. The errors in the Tables are random standard errors only. These are derived from the experimentally-measured standard deviation of individual spark counter results when counting different films from equally-irradiated dosimeters, and therefore include the statistics in the fission process (Harrison et al., in preparation).

Average fluence to dose-equivalent conversion factors for the neutron sources (Table I, col. 3) were calculated using the spectra described in the preceding sections and conversion factors given by the ICRP (ICRP, 1973). ^{237}Np (n,f) cross-sections from Delafield et al. (1973b) were used to derive the effective theoretical cross-sections (Table I, col. 7). Only for the Pu-Be source were other published values used (Burger et al., 1971). Errors in the neutron fluence measurements and in the spectrum-averaging procedures for the cross-sections and conversion factors are thought to be each $\lesssim 5\%$.

The average conversion factors were used to derive the dose-equivalent responses of the dosimeters (Table I, cols. 4 and 5) and, together with the effective cross-sections, to obtain theoretical free-air dose-equivalent responses (Table I, col. 8).

The ratio of the experimental spark counts to the theoretical number of fissions was then calculated for each source to see if consistent values were obtained for the track detection efficiency of the spark counter (Table I, col. 9).

The variation of the albedo response with the divergence of the neutron beam was investigated using the ^{252}Cf source. However, the albedo response for this source was found to be very small at all distances (Table II), so the result is inconclusive.

Angular-dependence measurements with the ^{252}Cf source (Table II) and the Pu-Be source (Table III) showed no statistically significant angular variations except for the Pu-Be source where a small difference in the 0° - 180° sensitivity of the dosimeter in free-air was observed. This is in agreement with the observations of Mijnheer (1974) for higher-energy neutrons.

For Pu-Be neutrons the ratio of the response of the dosimeter on the front of the phantom (at 27 cm from the source) to the response when the dosimeter was on the back of the phantom was found to be approximately 6:1, when the distance between the source and dosimeter was kept constant.

3.7 Discussion

With the exception of the thermal-neutron response, the cross-sections tabulated by Delafield et al. (1973b) give a good description of the free-air response of a neptunium dosimeter if a plausible energy-dependent spark-counter efficiency is assumed. This efficiency is measured to be apparently about 0.5 at low neutron energies up to 1 MeV, then increases steadily up to 0.7 at 4 MeV. Such an increase is qualitatively but not quantitatively consistent with the forward-peaking in the angular dependence of neutron-induced fission in neptunium (Androsenko et al., 1969), which would predict a maximum variation of much less than 25% (see Section 2.4). This lesser value would be supported by the relatively slight angular dependences seen for the dosimeter with the high-energy neutron sources. Since the high-energy cross-sections are well-substantiated (see Section 2.2), this may imply that the low-energy cross-sections used are too high by between 20 and 40%, and the corresponding spark-counter efficiency should be between 0.6 and 0.7 at low energies. The anomalous efficiency deduced for thermal neutrons (0.82) is misleading, and can probably be explained by slight contamination of the neptunium by an isotope with a high thermal-neutron fission cross-section (see Section 5.1).

The sub-threshold response in dose-equivalent units is clearly much higher than has been anticipated previously (Cross and Ing, 1975). In particular, the free-air result for the moderated Sb-Be source shows a dose-equivalent sensitivity very similar to that for the fast neutron sources. This is however coincidental as the dosimeter is relatively insensitive to neutrons between 10 keV and 375 keV and must therefore be oversensitive to those below 10 keV. This is consistent with the response illustrated in Fig. 1.

Because of this significant response to low-energy neutrons there is a marked sensitivity to albedo neutrons, when the dosimeter is irradiated on a phantom. This is of greatest importance in the region of neutron energies from 1 keV to 100 keV, where the free-air response alone is quite small. In a separate paper (Gibson, in preparation), the on-phantom results are compared with a calculation based on new theoretical albedo-neutron spectra.

The low response of the dosimeter to neutrons incident through the back of the phantom, even for the relatively penetrating Pu-Be neutrons, indicates that its sensitivity when worn on the body is substantially confined to the forward hemisphere.

4. Measurements of Possible Interfering Effects in the Track Detector

Interfering effects in the polycarbonate fission-fragment detector could be produced by the high doses of α , β and γ radiations which it receives in prolonged contact with the neptunium (and its β -emitting daughter protactinium). These could, for example, lead to changes in the etching characteristics of the plastic, and spurious background counts. In addition, there may be some detectable response to heavy-ion recoils produced by neutrons, and the latent fission-fragment tracks themselves may be subject to fading prior to etching. Published literature (see references below) suggests that none of these effects will produce a significant problem in the proposed personal dosimeter. However, some additional experiments were undertaken to investigate possible interferences, and these are described in the following sections (4.1-4.3).

4.1 Sensitivity to α , β and γ radiations

The absence of sensitivity of polycarbonate to α , β and γ radiations at doses up to about 10 Mrad has been widely reported (Becker, 1972; Fleischer et al., 1975). To confirm these reports, samples of polycarbonate film were irradiated with doses of up to 0.4 Mrad of α radiation from a ^{241}Am source, 1 Mrad of β radiation from a ^{90}Sr source and 5 Mrad of ^{60}Co γ radiation. No spark counts were recorded in any case after normal processing, and the plastic was not sensitive to 3 MeV or 5.5 MeV α particles even after these high doses of β and γ radiation. This is of some importance because the polycarbonate film in contact with 0.5 mg cm^{-2} ^{237}Np is irradiated by α particles at a rate of about $0.3 \text{ Mrad month}^{-1}$.

4.2 Sensitivity to heavy ion (C,N,O) recoils

Polycarbonate has been shown by microscopic examination to be sensitive to C, N and O recoils (Becker, 1972). However, these tracks are very short ($1\text{--}4 \mu\text{m}$) and should not be counted by the spark counter because they do not etch through the plastic. Samples of polycarbonate sandwiched between teflon or polythene were irradiated with large doses of Pu-Be neutrons equivalent to about 200 rem. Only one or two tracks were subsequently counted by the spark counter after standard processing. Direct recoil registration is therefore not a significant source of neutron sensitivity in this detection system.

4.3 Fading of latent tracks

It has been reported by Becker (1973) and Sayed and Piesch (1974) that the fading of latent fission-fragment tracks in polycarbonate is very slight in dry conditions. However, Sayed and Piesch (1974) also report that in 97% humidity nearly 20% fading occurs after one month at 50°C , while Becker (1973) detected no loss of tracks in $10 \mu\text{m}$ polycarbonate film stored for 3 months at 30°C in 95% relative humidity.

To substantiate these reports, sample polycarbonate films were irradiated and stored on a radiator in the laboratory in normal daylight at between 35° and 45°C for one and two months before etching. No fading occurred in one month (within a likely statistical error of $\pm 4\%$), but after two months in the same conditions $9 \pm 4\%$ fading was observed. In controlled-humidity experiments at lower temperatures, no fading was detectable in films stored for one and two months at 25°C and constant 56% or 86% relative humidity. However, films etched and counted after being stored in a desiccator at 25°C for one or two months all showed an unexpected increase in sensitivity of about 10%.

It can be concluded therefore that, on a timescale of one or two months, fading or enhancement is unlikely to be as much as $\pm 10\%$ under normal environmental conditions.

5. Design and Safety Criteria

The first requirement in the design of a neptunium personal dosimeter is to ensure that the minimum of radioactive neptunium of sufficient purity is used. The neptunium must be contained as well as possible and the penetrating radiations emitted by it must be adequately shielded from the wearer. Finally, a security system must be employed to avoid the loss of dosimeters. These requirements are discussed in the following sections (5.1-5.4).

5.1 Quantity and purity of ^{237}Np

In order to obtain a practical sensitivity of about 1 count per 5 mrem, 4 mg (2.8 μCi) of ^{237}Np is required in the form of a thin source for fission-fragments.

The neptunium must be free from other fissionable isotopes. An impurity level of 15 ppm of an isotope having a large thermal-neutron fission cross-section (principally ^{235}U and ^{239}Pu) would give an enhancement of about 50% in the response to thermal neutrons, and a slightly increased albedo-neutron response up to about 10 keV neutron energy. Such an impurity level would be just tolerable. Isotopes with negligible thermal-neutron fission cross-sections (< 100 mbarns) may be present in concentrations up to 10^3 ppm, but it is vital that very high spontaneous-fission rate isotopes such as ^{252}Cf are absent as even in very small concentrations (parts in 10^{12} - 10^{14}) they can introduce unacceptably high background fission rates in the dosimeters. The best method for measuring these impurity levels is the direct one of measuring the response of the dosimeter to a calibrated thermal-neutron flux, if necessary by a difference method using a borated shield (section 3.1), and by measuring the spontaneous-fission rate using unirradiated dosimeters.

These experiments have been undertaken for a sample of "high purity" neptunium obtained from The Radiochemical Centre Ltd., Amersham, UK, and values of 33 mbarns and $1.0 \text{ fissions mg}^{-1}\text{month}^{-1}$ obtained for the thermal-neutron cross-section and the spontaneous-fission rate, respectively. These values are 65% and six times higher respectively than would be expected for pure neptunium (Lederer et al., 1967), but are acceptable for the proposed dosimeter. The spontaneous-fission rate measurement almost certainly contains a neutron-induced or photofission component from natural and environmental background radiation, and therefore represents an upper limit to the true spontaneous-fission rate.

5.2 Containment of the fissile material

The best technique for obtaining an adherent layer of neptunium about 0.5 mg cm^{-2} in thickness on stainless-steel, platinum or aluminium is to paint on a solution of neptunium nitrate in ethanol and to "burn-in" the deposit (Sohrabi and Becker, 1972). The source can then be sealed into a suitable metal or plastic holder behind a thin window which serves as additional protection against the transfer of radioactivity by contact. A simple sandwich of the source and window between two pieces of aluminium bonded together with Araldite adhesive is proposed for this purpose (see Fig. 3).

Various window materials have been tried for covering the neptunium. Nickel or plastic materials seem to be the most suitable. $660 \mu\text{g cm}^{-2}$ Ni foil is easy to handle, but is quite fragile and is attacked by chemicals. Polycarbonate is a tough radiation-resistant plastic which is available commercially in thicknesses down to $1.8 \mu\text{m}$. Polycarbonate $2 \mu\text{m}$ thick, with a thin aluminized coating, is available for X-ray proportional-counter windows (Siemens Ltd.) and this is easier to handle than the plain material, which is usually supplied on a backing. Tests showed that the plastic did not deteriorate after irradiation by the equivalent of seven years of alpha-radiation from 0.5 mg cm^{-2} neptunium source (simulated using a ^{241}Am source). The effective transmission of the aluminized material for fission-fragments was measured using the spark counter to be about 80%.

5.3 Radiation shielding of the neptunium

^{237}Np decays by alpha-particle emission ($t_{1/2} = 2.1 \times 10^6 \text{ a}$) to ^{233}Pa , which itself decays by β particle emission with a short half-life (27 days) to ^{233}U . This has a long half-life ($1.6 \times 10^5 \text{ a}$). ^{233}U is an undesirable isotope because it has a large thermal-neutron fission cross-section (524 barns). However, its concentration does not reach 15 ppm until 46 a after the neptunium has been purified.

The gamma-ray spectrum of a ^{237}Np source contains a number of ^{237}Np and ^{233}Pa lines, of which the most intense are 86.5 keV, 94.6 keV, 98.4 keV, 300.1 keV and 311.9 keV. Calculations using tabulated absorption cross-sections (McMaster et al., 1969) showed that 3 mm of lead reduces the dose-rate from all the lines below 100 keV to < 1 % of the dose-rate due to the 311.9 keV line. This line is itself attenuated to about 28% of its value with no radiation shield. These values were checked by gamma-ray spectrometry. The build-up of characteristic X-rays from lead is unimportant at this thickness. A 3 mm tungsten shield is even more effective, reducing the gamma-ray at 311.9 keV to 18%. However, these thick shields also perturb the neutron flux and lead is preferable to tungsten because it has a lower scattering cross-section at all neutron energies, and no resonant structure (Hughes and Harvey, 1955).

The surface dose-rate to the wearer using a 3 mm lead shield and 3 mm of plastic was measured using a stack of thin thermoluminescent dosimeters to be 11 mrem/40 h. The dose is localized to the region of the body below the dosimeter, but the radiation is quite penetrating so that the dose-rate behind 1 g cm^{-2} of tissue under the dosimeter is about 8 mrem/40 h. However, the dose-rate to the male gonads for a dosimeter worn at waist level is about $1 \mu\text{rem h}^{-1}$ (2 mrem per working year). For a female, the gonad dose-rate is slightly less, as the closer proximity of the gonads to the dosimeter is more than offset by the shielding provided by the tissue and other organs of the body.

An exploded view of the proposed dosimeter incorporating a 3 mm lead radiation shield is shown in Fig. 3.

5.4 Safety procedures

In spite of the containment of the source and the radiation shield, it is undesirable that dosimeters containing radioactive sources should be taken away from the radiation areas where they are used. By fixing the dosimeter to a belt which has to be removed when leaving the radiation area, such occurrences would be made much less likely. Furthermore, the dosimeter could be sealed onto the belt to discourage unauthorized attempts to dismantle it.

At the end of the issue period, the entire belt would then be returned to the Health Physics staff, who would replace the polycarbonate film in the dosimeter and re-seal it, ready for re-issue.

6. Conclusions

The ^{237}Np fission dosimeter offers a replacement for the nuclear track emulsion which is both easier and quicker to interpret, covers a wider range of neutron energies and is less subject to errors. Its response when worn on the body is closer to dose-equivalent over the whole range of neutron energies than any other type of neutron dosimeter currently available. The capital cost of the dosimeters is likely to be very much higher than the track emulsion, but the negligible cost of consumable materials and lower manpower commitment to read the dosimeters would soon recover the outlay.

The dosimeter suffers from the major drawback that it contains radioactive material and the wearer is subject to a very small localized radiation dose during use. Such a dosimeter should therefore only be used where the security is adequate, and the radiation hazards are real.

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Table I
The Experimentally-measured Response of the ^{237}Np Dosimeter to Neutrons

Neutron Source (1)	Neutron Energy (mean) (2)	Conversion Factor ($\text{n cm}^{-2}\text{rem}^{-1}$) (3)	Experimental Response			Calculated Response		Apparent Counting Efficiency Ratio $\frac{(4)}{(8)}$ (9)
			Free-air (counts rem^{-1}) (4)	On-phantom (counts rem^{-1}) (5)	Ratio $\frac{(5)}{(4)}$ (6)	Fission Cross-section (barns) (7)	Free-air Sensitivity (fissions rem^{-1}) (8)	
GLEEP	thermal	9.36×10^8	156 ± 13	(281)†	(1.8)	0.020	191	0.82 ± 0.07
moderated Sb-Be	~ 0.5 keV	6.37×10^8 †	313 ± 20	410 ± 100	1.31 ± 0.33	$0.083^\dagger$	539	0.58 ± 0.04
Sb-Be	23 keV*	5.40×10^8	39 ± 5	183 ± 21	4.69 ± 0.81	0.014	77	0.51 ± 0.06
IBIS	0.10 MeV	1.73×10^8	(16)**	50 ± 10	(3.1)**	0.018	32	(0.49)
"	0.24 MeV	8.28×10^7 †	19 ± 2	30 ± 3	1.58 ± 0.23	$0.050^\dagger$	42	0.45 ± 0.05
"	0.55 MeV	4.50×10^7 †	152 ± 6	193 ± 7	1.27 ± 0.08	$0.63^\dagger$	289	0.53 ± 0.02
"	1.12 MeV	2.95×10^7	223 ± 5	260 ± 6	1.17 ± 0.04	1.50	451	0.49 ± 0.01
"	1.72 MeV	2.63×10^7	241 ± 5	279 ± 12	1.16 ± 0.06	1.62	435	0.55 ± 0.01
^{252}Cf	2.13 MeV	2.95×10^7 †	260 ± 6	266 ± 5	1.02 ± 0.03	$1.31^\dagger$	394	0.66 ± 0.02
Pu-Be	4.04 MeV	2.77×10^7 †	293 ± 11	314 ± 11	1.07 ± 0.05	$1.48^\dagger$	418	0.70 ± 0.03

*375 keV component subtracted - see text

†Spectrum-averaged values - see text

**Theoretical free-air value (using an interpolated spark counter efficiency of 0.49)

†Assumes a thermal-neutron albedo of 0.8

Table II

The Experimental Directional and Distance Dependence of the Response of the Dosimeter to ^{252}Cf Neutrons in Free-air and On-phantom

Orientation of Dosimeter*	Source-dosimeter Separation (cm)	Relative Response Free-air	Relative Response On-phantom
0°	15	1.00	1.00
45°	15	1.00 ± 0.03	0.98 ± 0.04
180°	15	0.96 ± 0.03	-
0°	20	0.98 ± 0.03	0.97 ± 0.02
0°	35	1.03 ± 0.04	1.06 ± 0.03
0°	70	1.02 ± 0.07	1.00 ± 0.05

*0° orientation is defined by the neptunium layer being normal to neutron beam, with the track detector behind

Table III

The Experimental Directional Dependence of the Response of the Dosimeter to Pu-Be Neutrons in Free-air and On-phantom

Orientation of Dosimeter†	Source-dosimeter Separation (cm)	Relative Response Free-air	Relative Response On-phantom
0°	15	1.00	1.00
45°	15	0.99 ± 0.03	1.07 ± 0.04
180°	15	0.93 ± 0.03	-

†defined as in Table II

A.E.R.E. R8679 FIG. 1.

AERE - R 8679 Fig. 1

The theoretical dose-equivalent sensitivity of ²³⁷Np to neutrons (free-air)

A.E.R.E. R8679 FIG.2.

AERE - R 8679 Fig. 2

The theoretical dose-equivalent sensitivity of ²³⁷Np to gamma radiation (free-air)

AERE - R 8679 Fig. 3
Exploded view of the proposed dosimeter