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A DOSEMETER BASED ON NUCLEAR ETCHED TRACK DETECTORS FOR THERMAL, FAST AND HIGH ENERGY NEUTRONS WITH FLAT RESPONSE

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Abstract — The response of a neutron dosemeter based on plastic track detectors has been studied. The dosemeter (converter + detector) configurations used consist of a CR-39 detector 500 μm thick and a Makrofol converter 300 μm thick for fast neutrons (configuration 1) and 3 mm of air used as converter for thermal neutrons (configuration 2). The possibility of using Makrofol as a high energy neutron dosemeter has also been studied. In order to validate the results obtained from Monte Carlo simulations using the MCNP4A code, a set of irradiations by monoenergetic neutron beams has been performed at the Physikalisch-Technische Bundesanstalt (PTB), Gesellschaft fur Strahlen-Und Umweltforschung (GSF) and Paul Scherrer Institut (PSI) neutron irradiation facilities. Irradiations with realistic fields were performed at the Cadarache Nuclear Centre. An excellent agreement has been found between the simulated and the experimental values, not only for fast neutrons but also for thermal neutrons.

INTRODUCTION

The ICRP 60 recommendations[1] reduce from 50 mSv to an average of 20 mSv over 5 years the annual dose limit for occupational exposure of the whole body, establish the required minimum detectable dose of a detector to 80 μSv for monthly issue corresponding to the recording level of 1 mSv and increase the weighting factor for fast neutrons up to 100% depending on neutron energy.

Despite the great progress achieved during recent years in the field of neutron dosimetry as a consequence of the addition of bubble detectors[2,3] and silicon diodes[4,5], it may be stated that there is no dosemeter able to measure, with adequate accuracy, neutron dose independently of the neutron spectrum.

When studying the response of a given dosemeter, the influence of the etching conditions must be taken into account. The purpose of this work is to characterise the response and the detection limit of a neutron dosemeter, based on nuclear etched track detectors and designed in our laboratory, with the aim of fulfilling the ICRP 60 requirements, when a two-step electrochemical etching (ECE) procedure at low and high frequency is used.

The study of response was performed using the MCNP4A[6] code and the PROT program[7] and validated with a series of irradiations to monoenergetic neutrons (PTB, GSF and PSI) and realistic neutron fields (Cadarache)[7].

EXPERIMENTAL PROCEDURE

Dosemeter arrangement

Configuration 1 is based, following the direction of incidence of the neutron beam, on a 3 mm thick layer of polyethylene followed by 300 μm of Makrofol-ED polycabonate (manufactured by Bayer AG), which acts both as converter for fast neutrons and as detector, on a 500 μm thick layer of PADC (CR-39, manufactured by Pershore Moulding Ltd with a curing time of 32 h), and on a 5 mm thick methacrylate holder. To increase the sensitivity of this configuration to thermal neutrons, 3 mm of air has been added to this configuration, following the Makrofol layer, which acts as the converter for thermal neutrons using the reaction $^{14}N(n,p)^{14}C$: this configuration will be called configuration 2. To study the albedo contribution, two more configurations have been made (3 and 4) from the second one adding 1 mm of cadmium in front of the polyethylene (configuration 3) and behind the methacrylate holder (configuration 4).

All dosemeter configurations were sealed into Climafol pouches. These pouches, which are composed of 12 μm of polystyrene, 12 μm of aluminium and 75 μm of polyethylene were taken into account for simulation, as well as the phantom[8].

In the simulations, the energy used was the same as that for the experimental irradiations, and the fluence was 10^6 neutron.cm^{-2}. The surface of the dosemeter was considered to be 1 cm^2 and the surface of the phantom, 25 cm^2, which ensured that the ratio between these surfaces is equal to that between the surfaces of the real dosemeter and phantom.

For irradiation, the dosemeters were put together in 81 cards of 6×6 cm^2 after determining the side of the CR-39 which presents the lowest background. Each irradiation card was sealed inside a Climafol pouch and stored inside a refrigerator until irradiation.

Neutron irradiations

Sixty cards of configuration 1 were irradiated during the Eurados-Cendos joint irradiation experiment in 1992

at the GSF (Neurenberg) and PTB (Braunschweig) to a range of neutron dose equivalent between 1.5 and 3.5 mSv of monoenergetic beams of 144 keV, 565 keV, 1.2 MeV, 5.3 MeV, 15.1 MeV, as well as to neutrons from a ^{252}Cf source. Irradiations were performed at 0° (normal incidence), 30°, 60° and 85° incidence. Three cards were irradiated at the PSI (Villigen) to high energy neutrons, in fields with broad spectral distributions with maxima at 44 MeV and 66 MeV, with an average energy of 25 MeV and 38 MeV, respectively[9].

Twelve cards, one for each configuration and source, were irradiated during a joint irradiation experiment in 1995[10], at the LRDE in Cadarache, at different realistic neutron fields (Sigma, Canel+, Canel+ plus water)[11].

Three irradiation cards for monoenergetic neutrons, together with six cards for realistic neutron fields which correspond to configurations 1 and 2, were kept together with those to be irradiated to study the background during transit.

After irradiation and before the etching process, each CR-39 and Makrofol card was cut into nine 2×2 cm^2 plates.

Processing and reading

The etching device consists of 10 cells[7], each one for two detectors. All samples of each irradiation energy set and a background detector were etched simultaneously. All the plates were etched in our electrochemical etching system[7], and the cells containing the detectors and the etching solution are separately kept overnight at the etching temperature. The etching solution is placed inside the cells at the moment the etching starts.

The CR-39 foils were etched using 6N KOH aqueous solution at 60.0±0.1°C, with the following etching steps:

1st step: 20 kV.cm^{-1} RMS at 50 Hz for 5 h,

2nd step: 20 kV.cm^{-1} RMS at 2 kHz for 1 h,

3rd step: 15 min post-etching.

The Makrofol foils were etched in a similar process using a mixture of 6N KOH and 40% ethanol at 35°C and 26.7 kV.cm^{-1}.

The tracks were counted by means of an image analysis system based on Visilog 5.1 coupled to a photo video camera Sony PAV-A7E in an area of 0.69 cm^2. The reading of the CR-39 and Makrofol foils was not corrected for field strength dependence and dose linearity[12].

The dose equivalent response in terms of H*(10) has been evaluated from net measured track densities (measured track density minus average background). The response in terms of H′(10,α) is calculated from H*(10) by means of the appropriate conversion factors[13,14].

RESULTS AND COMMENTS ON THE DOSEMETER IN VARIOUS FIELDS

Energy dependence

For the configurations 1 and 2, the simulated and the measured directional dose equivalent response $R_{H'}$ (ratio between the net track and the reference ambient dose equivalent) for CR-39 (shaded points) and Makrofol (unshaded points) for normally incident neutrons is shown in Figure 1 as a function of neutron energy from thermal up to 66 MeV. As can be seen, there exists a good agreement between the simulated (solid line) and experimental results, finding little difference in the responses $R_{H'}$ corresponding to configurations 1 and 2 for neutrons with energy greater than 144 keV[15]. The variation of the response $R_{H'}$ for the CR-39 was smaller than 40%, a value very acceptable if we take into account the neutron energy range studied. The response of the Makrofol detector in the region up to 15.1 MeV was 50% lower than that corresponding to CR-39, and its variation in the complete range studied (^{252}Cf–66 MeV) was of about 40% too. Our experimental values are in good agreement with those published by Piesch *et al*[16] and Luszik-Bhadra *et al*[17] except for the energies of 46 MeV and 66 MeV. In addition, we do not have simulated results for these energies, as they are higher than the limits allowed by MCNP4A code.

In Table 1 we present the experimental and simulated directional dose equivalent response $R_{H'}$ for the three realistic neutron beams. The excellent agreement between the simulated and experimental results for all the configurations must be emphasised.

The values found for configurations 1 and 2 in these

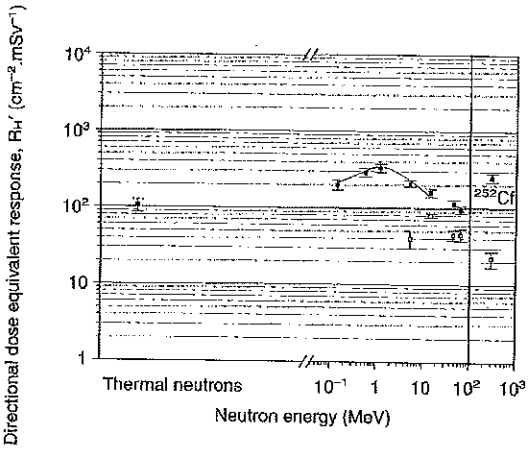

Figure 1. Experimental and simulated directional dose equivalent response, $R_{H'}$, for normally incident neutrons as a function of neutron energy from thermal up to 66 MeV. (■) Experimental values for CR-39, (□) experimental values for Makrofol, (— —) simulated values.

three cases, are self-consistent if we take into account that the contributions to the response from thermal and fast neutrons are of 40% and 60% for Sigma, 7% and 90% for Canel+ and iron and 15% and 60% for Canel+ and iron and water.

The very similar values of the responses found in the results regarding configurations 2 and 4 is due to the fact that the albedo component, which in a normal situation would appear as the difference between configuration 2 and 4, decreases as we have seen in the simulation, while going through the different elements which involve configuration 2, being practically zero when it arrives at the air converter.

The directional dose equivalent response values for normal incidence in configurations 1 and 2 were $(275\pm50)\ cm^{-2}.mSv^{-1}$ and $(130\pm20)\ cm^{-2}.mSv^{-1}$ respectively.

Angular dependence

For configuration 1 of the dosemeter, the simulated and experimental directional response $R_{H'}$ for monoenergetic neutrons of 144 keV, 565 keV, 1.2 MeV, 5.3 MeV, 15.1 MeV as well as for neutrons originated by a ^{252}Cf source are displayed in Figure 2 for CR-39 and Makrofol as a function of the neutron incidence angle. A very good agreement is observed between the simulated and experimental results, but showing a rather poor angular response dropping by a factor of about 2 from the response at 0° to 60° incidence.

In order to use the dosemeter in routine measurements, a calibration factor for all neutron incidence and energies has been calculated as the reciprocal of the mean directional dose equivalent response $R_{H'}$ to 0°, 30° and 60° neutrons from ^{252}Cf. The per cent deviation between the directional dose equivalent response using the calibration factor obtained, $1/(197\pm86)\ cm^{-2}.mSv^{-1} = (5.08\pm2.20)\times10^{-3}\ cm^2.mSv$, and the real dose values ranges between +60% and −40% except for the points at 144 keV and 15.1 MeV at 60°. These results are in very good agreement with those obtained by Luszik-Bhadra *et al*[12,18].

Detection limit

The detection limit L_D, defined as the lowest dose that can be detected with a specified level of confidence (usually 95%), has been obtained for all energies and incidence angles according to Christensen and Griffith[19]. For CR-39 and neutron incidence in the range 0°–60° a value of 60 μSv has been found for all energies except for 144 keV at 60°, which is below the 80 μSv limit recommended by the ICRP 60. This low value is the consequence not only of having CR-39 with good manufacturing conditions, but also of the careful manipulation during all the making, irradiation and etching of the CR-39 detector. In addition, the fact that detectors have been stored in sealed Climafol pouches ensures that the exposure to radon and radon daughters has been minimised.

The value found for the detection limit in the Makrofol was about six times greater.

CONCLUSIONS

From the results of this work, it can be stated that the computer code allows the experimental results to be reproduced with excellent agreement. It is possible to define the most adequate dosemeter (converter +detector) geometry for thermal and intermediate neutrons if the cross sections of the appropriate nuclear reactions are introduced in the code. The code can also be extended in order to account for reactions involving neutron collisions with converter or detector C and O nuclei, which may be relevant for high energies as they originate alpha particles that could be recorded and that, consequently, may affect the dosemeter response.

The dosemeter with configuration 2 presents an acceptable energy response, even though the value $150\pm20\ cm^{-2}.mSv^{-1}$ of directional dose equivalent represents 54% of the $275\pm50\ cm^{-2}.mSv^{-1}$ of configuration 1. The results of the simulation allow us to state that a thickness of 4 mm of air will be enough to equalise the response of both configurations. In this way, a dosemeter (configuration 2) is obtained that is able to

Table 1. Experimental and simulated directional dose equivalent response $R_{H'}$ for realistic neutron beams (Sigma, Canel+ with iron and Canel+ with iron and water) and normally incidence neutrons.

Dosemeter configuration	Sigma		Canel + plus iron		Canel + plus iron and water	
	Experimental results ($cm^{-2}.mSv^{-1}$)	Simulated results ($cm^{-2}.mSv^{-1}$)	Experimental results ($cm^{-2}.mSv^{-1}$)	Simulated results ($cm^{-2}.mSv^{-1}$)	Experimental results ($cm^{-2}.mSv^{-1}$)	Simulated results ($cm^{-2}.mSv^{-1}$)
1	110±15	105±10	275±50	290±15	130±15	145±10
2	130±25	115±10	295±35	275±15	150±20	150±10
3	85±25	95±10	280±35	285±15	135±15	135±10
4	75±20	115±10	310±55	275±15	140±25	150±10

provide a flat directional dose equivalent response in a very large range of energies, except for the intermediate and albedo neutrons for which this configuration is insensible. In addition, deviations from the true value of directional dose equivalent response when it is evaluated using the calibration factor obtained in this work, ranges between +60% and −40%, except for the two particular points at 144 keV and 15.1 MeV at 60°. The detection limit of the dosemeter has been found to be 60 μSv, due mainly to the careful manipulation during all the making, irradiation and etching of the CR-39 detector.

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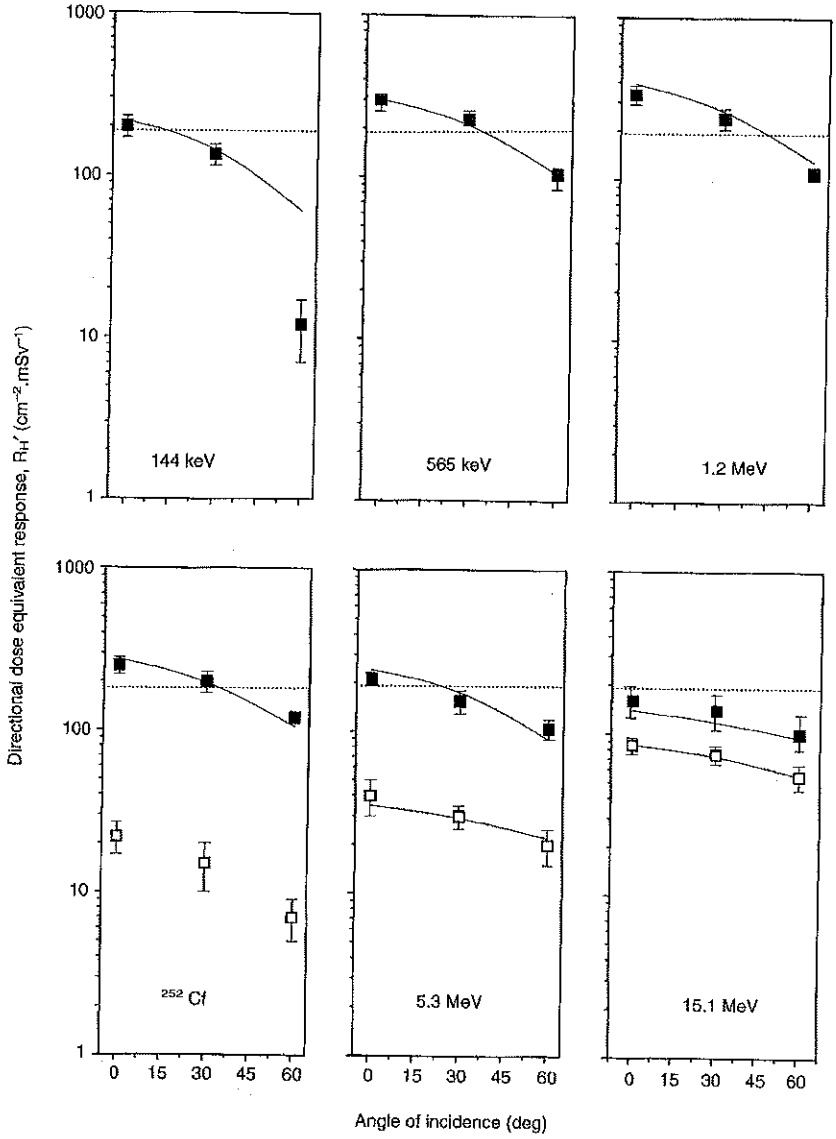

Figure 2. Experimental and simulated directional dose equivalent response $R_{H'}$ as a function of neutron angle of incidence for several monoenergetic neutron beams and ^{252}Cf source. (■) Experimental values for CR-39, (□) experimental values for Makrofol, (——) simulated results. The value used for calibration factor is indicated with a dashed line.

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