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THE PERFORMANCE OF A FISSION TRACK DOSIMETER FOR MONITORING THE NEUTRON RADIATION FROM ^{252}Cf SOURCES ON HANDS INSIDE GLOVE BOXES

by

B.J. MIJNHEER

1973

Joint Nuclear Research Centre
Geel Establishment - Belgium

Central Bureau for Nuclear Measurements - CBNM

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The angular dependence of the response of the dosimeter with thorium and other fissile materials was measured at different energies.

The neutron sensitivity of the dosimeter was determined with a calibrated ^{252}Cf source and amounts to 14.9 $\pm$0.7 mrem. spark^{-1}.

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KEYWORDS

MONITORING
DOSEMETERS
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CALIFORNIUM 252
RADIATION SOURCES
GLOVE BOXES
PERSONNEL DOSIMETRY

CONTENTS

1. Introduction

For the preparation of fission and neutron sources microgramme quantities of ^{252}Cf are handled at CBNM in a water shielded glove box with inner dimensions: height 70 cm, width 80 cm and depth 70 cm, with a water layer of 20 cm between 2.5 cm thick Plexiglas walls at the bottom, the front and both sides. The presence of the water shield will protect the body of the operator outside the box, but will simultaneously increase the dose equivalent on the hands due to scattered neutrons, scattered gamma-rays and gamma-rays from the capture of thermal neutrons by hydrogen. It was therefore decided to wear neutron dosimeters on the hands for monitoring the neutron dose equivalent, whereas TLD-100 chips should be worn in order to measure also the gamma dose.

At this moment a personnel neutron dosimeter with a sensitivity which is over a large neutron energy range proportional to the dose equivalent curve (the conversion factor of the flux density to the dose equivalent rate as a function of the neutron energy) does not exist. The best thing one can do therefore is to measure the neutron spectrum at the place to be monitored and to select the personnel neutron dosimetry system that is the most suitable for his situation. For work outside the shielding of reactors albedo type dosimeters are suitable (1), (2). These dosimeters have a constant dose equivalent response over the energy range from thermal to about 10 keV. Because of this upper limit the use of the nuclear track emulsion dosimeter can be advantageous in some situation e.g. during work with unshielded or slightly moderated $Be(\alpha, n)$ sources and ^{252}Cf sources (3).

However, the disadvantages of the nuclear emulsion dosimeter are well-known (4) and the advantages of a neutron

dosimeter based on fission track registration were soon realized by Walker, Price and Fleischer (5) since their discovery that tracks from heavy nuclear particles in certain insulators could be observed visually. Counting of fission tracks in thin plastic foils has become rather easy since the introduction of the electrical track counter (ETC) by Cross and Tommasino (6) , and succesful replacement of the nuclear track emulsion dosimeter by personnel neutron dosimeters based on fission track registration, has been reported by several authors [e.g. (7), (8)].

Due to their dimensions, wearing these dosimeters on hands is not very suitable, and a special thorium dosimeter to be attached to a finger tip (9) and a ^{237}Np dosimeter worn as a fingerring (4) are reported. These dosimeters are rather small and will thus have a rather low sensitivity. Also touching the thin plastic foil during the loading of the dosimeter, the etching and counting, may cause damages. Preliminary measurements with TLD chips on the finger tips and on the back of the hand, showed that inside a glove box the gamma dose on the hand is of the same order of magnitude as the dose on the finger tip. The same observation was made for the radiation doses to the wrists and fingers of workers engaged on radio-chemical processing in glove boxes (10). Though this ratio may be different for the neutron dose equivalent, it was nevertheless decided to make a fast neutron dosimeter based on fission track registration for wearing on the back of the hands. Such a dosimeter should be made rather large in order to enhance the sensitivity, should be easy to assemble and disassemble without touching the thin plastic foil and should have a zero background. Addition of a thermal neutron dosimeter, e.g. a ^{235}U fission track dosimeter, did not seem very useful as the dose equivalent due to thermal neutrons was expected to be very small.

As the excitation curve of the Th(n, f) reaction does not follow the dose equivalent curve, one has to know the neutron spectrum in which the Th-dosimeter has been irradiated. If the neutron spectrum is known, e.g. in the case of a pure ^{252}Cf spectrum, and the Th-dosimeter has been calibrated in such a spectrum with a known flux density, then the dosimeter can be used on places where the same spectrum exists, in order to measure the dose equivalent. Inside the glove box the neutron spectrum will, however, deviate from a pure ^{252}Cf spectrum due to the presence of scattered neutrons, and a determination of the neutron spectrum is necessary to obtain the total neutron dose equivalent.

The hand will absorb neutrons, and a dosimeter put on the back of the hand will only measure the neutron fluence at the position of the dosimeter. Knowledge of the average neutron fluence over the whole hand, which can be converted to neutron dose equivalent by means of whole body fluence to dose equivalent conversion factors, will be difficult as the orientation of the hand towards the source will vary. Also there will in general be more than one neutron source present in the box, or the box may be contaminated. In practice it is therefore easier to convert the fluence measured at the back of the hand to dose equivalent.

During the handling of ^{252}Cf needles for therapy purposes behind a polythene shield in the neighbourhood of a patient, about the same shielding circumstances exist as in a glove box. The dosimeter will thus also be very useful for wearing on the hands during implantation work. Multiple scattering of neutrons may be somewhat less important in this case, depending on the type of shield. As these heavily moderated neutrons contribute only to a small percentage to the extra dose equivalent, the same sensitivity may be assumed for the dosimeter as is done for work inside a glove box.

. Description of the use of the dosimeter

.1. Composition of the dosimeter

The composition and dimensions of the final dosimeter are given in figure 1 and figure 2. As can be seen from figure 1, the dosimeter consists of two thorium discs (two instead of one in order to increase the sensitivity by a factor 2 and to reduce the directional dependence somewhat), that are in close contact with a Makrofol foil between them. The thickness of the thorium discs is 0.105 ± 0.005 mm, which is thicker than the maximum range of fission fragments in Th (< 15 µm). However, thinner thorium foils are less easy to handle, whereas the gamma dose rate from our dosimeter outside the stainless steel container was considered to be acceptable compared to the dose rate that normally exist inside a glove box. Flat thorium discs were fabricated from pieces of thick thorium at the Metallurgy department of CBNM. The discs, which had a diameter of 20 mm, were punched out of sheets with a thickness of about 0.1 mm, obtained by rolling the original material and annealing at a high temperature in vacuum. After polishing, the thorium discs were glued inside a 0.1 mm deep hollow part in the stainless steel container, in such a way that the surface of the thorium was at the same level as the surface of the container.

The gamma dose rate outside the stainless steel container was measured with calibrated TLD-100 discs. At the position of the Th-discs the exposure dose rate amounted to 1.05 ± 0.15 $mR.hr^{-1}$, whereas nearer the edge the dose rate dropped rapidly to 0.18 ± 0.07 $mR.hr^{-1}$. In the small box intended for the TLD chip, the exposure dose rate amounts to 0.09 ± 0.04 $mR.hr^{-1}$, indicating that small background corrections will be necessary if the TLD remains near the thorium over periods

longer than a few hours. As a result of preceding experiments stainless steel was chosen as the material for the container. Measurements of the γ-dose rate with TLD outside the dosimeter at the position of the thorium discs with containers of 1.0 mm perspex, aluminium and lead gave exposure dose rates of 4.0 ± 0.6, 2.0 ± 0.3 and 0.63 ± 0.06 mR.hr^{-1} respectively. The use of lead instead of stainless steel, does not lead to a considerable reduction of the dose rate, whereas the latter is much more suitable for making the dosimeter. If the dose rate is still considered too high and the weight of the dosimeter is not important, a lead foil might be added between the dosimeter and the hand.

Another possibility would be to reduce the amount of fissile material. A reduction of the thickness of the thorium foil to 10 μm will hardly influence the sensitivity of the dosimeter, whereas the exposure dose rate outside the stainless steel container at the position of the thorium discs will be reduced to 0.1 mR.hr^{-1}. However, the fabrication of such thin flat thorium discs will not be easy, and a special effort would be required to develop a technique for this purpose.

If the weight of the dosimeter with the stainless steel container (23.6 gram) is considered objectionable,than a perspex container can be used. This increases, however, the dose rate at the skin of the hand with a factor 4.

The handling of the 10 μm thick Makrofol KG foil can be simplified in the way described by Prêtre (11), by glueing the thin foil on a frame of 0.4 mm thick Makrofol E by means of a few small drops of chloroform. In this way the frame with the thin Makrofol foil can be placed in the dosimeter, etched and counted without touching the thin foil, thus avoiding any damage.

In the bottom part of the dosimeter a ring with a depth of 0.4 mm is hollowed out of the stainless steel, in which the Makrofol E frame fits precisely. By screwing together the two halves of the dosimeter, the thorium discs will be in close contact with the thin Makrofol foil. Before using the glued Makrofol KG foils, all foils were tested (counted with the ETC) to establish that no holes were present.

2.2. Etching

In the original version of the dosimeter 12 μm thick Makrofol KG foils were used, which were etched under circumstances as described elsewhere (12): one hour at (64 ± 1)°C in 6N NaOH. The background for these etching conditions amounted to 6 sparks per foil on the average, which was considered too high. As the background will be inversely proportional to the thickness of the counted foil, reducing the background can be obtained by using shorter etching times (13) or lower temperatures (7). Also the use of KOH instead of NaOH gave in the case of Lexan improvements in the background (14). It is, however, not certain that this will also be the case for Makrofol KG foil, as the etch rate of Lexan in a NaOH solution is strongly dependent on and that of Makrofol KG not at all dependent on etch products concentration (15).

For our final dosimeter with a 10 μm thick Makrofol KG foil, we used the etching conditions as given by Heinzelmann and Schüren (7): 75 minutes in a 35% in weight KOH solution in H_2O at 40°C. Background measurements as a function of the etching time showed that under these circumstances the background remained zero until 90 minutes etching time, while

for longer etching periods the background increased to about 3 tracks per foil at 150 minutes. To be on the safe side without lowering the efficiency too much, we maintained the etching time at 75 minutes. The background due to the spontaneous fission of ^{232}Th can be ignored.

2.3. Counting

After washing in distilled water and drying, the etched foils were counted by means of the ETC, an apparatus originally described by Cross and Tommasino (6). Figure 3 shows the ETC used at CBNM for counting the Makrofol foils from the dosimeters. The anode, made of copper on which a thin layer of nickel had been electroplated, was placed inside a hollow part of a piece of perspex. The frame with the thin Makrofol foil was then placed over this electrode in a small circular hollow in the perspex. A sheet of thin aluminized Mylar was pressed against the Makrofol foil and the other grounded electrode by means of a rubber covered piece of perspex just fitting the hollow part in the perspex containing the electrodes plus Makrofol foils.

Every foil was 8 times punched at 1100 Volt, after which the foil was counted 5 times at 550 Volt. A diagram of the electronic system is given in figure 4. The pulses coming from the ETC have been adapted to the counting device by means of a pulse shaping circuit, as given in the figure, and a discriminator.

For track densities smaller than 500 sparks per cm^2 the reproducibility of the counting was better than 1%. At higher track densities the reproducibility becomes worse. Also the linear relationship between the counted number of sparks and the number of emitted fission fragments, which is proportional to the neutron dose equivalent, no longer holds due to overlap of the holes in the aluminized Mylar foil.

Counting higher track densities in thin Makrofol foils can be done by means of a microscope. In this case one has to determine the detection efficiency of the microscope. If the counting through a microscope is not carried out visually but by means of an automatic image analyzer, than thin Makrofol foils can not be used, as it is difficult to discriminate between fission tracks and surface damage or α-tracks. If thicker plastic foils are used, good discrimination is possible, and in some types of dosimeters (9), (11), a sheet of thick Makrofol is therefore added in order to count with a microscope unusually high doses.

Another approach would be to modify the ETC in such a way that higher track densities can be counted. In a recent paper Cross and Tommasino (16) pointed out that the use of gases other than air may give sparks at lower voltages resulting in smaller holes evaporated in the thin Al-electrode, and thus decreasing the loss of counts due to overlap of holes at higher track densities. Experiments with our ETC using a 90% A/10% CH_4 gas mixture, showed that the starting voltage was lowered by about 50 Volt, but the plateau became shorter and had a steeper slope compared to that in air. Our result with He gas gave, contrary to the measurements of Cross and Tommasino a plateau at voltages higher than in air. Also the reproducibility was worse. The reason could be that we used a lower pressure of the He gas (1.5 atm. instead of 4.2 atm.).

According to other experiments by Cross and Tommasino (17) a change of the polarity of the Al-electrode has a large influence on the length and slope of the plateau count-rate versus high voltage, if thin (2.0 - 3.6 mm thick) plastic foils are used. However, for our foil thicknesses changing the

polarity had no observable influence on the plateau length (about 200V) and the slope (about 0.5% per 100V), nor did it influence the reproducibility of repeated counting the same plastic foil either.

More experiments will be necessary to improve our ETC in order to count higher track densities such as changing the construction of the ETC for the use of A/CH_4 gas at higher pressure. However, for the purposes for which we use the Th-dosimeter at CBNM, high doses are not to be expected, and the ETC in its present state will be satisfactory.

3. Angular anisotropy of the response of the dosimeter

3.1. Measurements with the thorium dosimeter and ^{252}Cf neutrons

One of the disadvantages of fast neutron dosimeters based on fission track registration, and which is also found in nuclear track emulsion dosimeters, is the dependence of the response on the angle between the impinging neutrons and the plane of the fission layer / plastic foil combination. This is due to the fact that fast neutron induced fission is strongly anisotropic. As the plastic foil is in contact with the fission layer, integration over a large range of angles will take place and the angular dependence of the dosimeter will therefore be smaller than that of the fission process itself.

Heinzelmann and Geiser (18), (19) have measured this effect with ^{232}Th and ^{238}U layers for different types of insulator foils, neutron energies and etching times for angles between 0° and 90°. The tracks were counted under the microscope. Their results showed a difference in response

of a factor 1.4 between irradiations carried out with the dosimeter perpendicular (90°) and parallel (0°) to the neutron beam. The anisotropy was only slightly dependent on the etching time, neutron energy and type of insulator foil. Pauw (13) noticed for thin fission layers in contact with 12 μm thick layers of Makrofol after etching for one hour at 64° in NaOH and counting with the ETC, a difference in response between 90° and 270°, which means irradiating the dosimeter with the front-side or the back-side towards the neutron source. His measurements showed that the effect was rather large at higher neutron energies, and it seemed therefore worthwhile to carry out angular anisotropy measurements with the Th-dosimeter in order to see whether the same effect occurred with thick layers of fissile material.

Preliminary measurements with a 1 mg ^{252}Cf source in the Euratom Transuranium Institute in Karlsruhe (Germany) showed only a small forward/backward effect. More accurate measurements at CBNM with a 10 μg ^{252}Cf source and irradiating two thorium foils simultaneously, showed a value of 1.008 ± 0.016 for the response ratio 270° to 90°. 12 μm thick Makrofol KG foils were used for these experiments, and were etched during 60 minutes at 60°C in a 6N KOH solution. Small corrections were made for distance differences between the two foils, whereas the correction for the difference in efficiency at different track densities ("overlap correction") was taken from Pauw (13) as the etching and counting conditions were the same. The effect is clearly small or absent for thick layers of Th, our foil thickness and our etching and counting conditions. The angular dependence for ^{252}Cf neutrons is given in figure 5, and shows that the shape is about the same as was measured by Heinzelmann for 14.5 MeV and AmBe neutrons and his etching and counting conditions.

3.2. Measurements with the thorium dosimeter and mono-energetic neutrons between 12.8 and 16.8 MeV

The theory of fission shows that near the thresholds of the reactions (n, nf) and (n, 2nf) etc., discontinuities can be expected in the anisotropy of fission, which may be rather large for even-even nuclei [Halpern and Strutinskii (20)]. Experimental data e.g. of Henkel and Brolley (21) for ^{232}Th supported these expectations. Measurements of the angular dependence of the dosimeter were therefore carried out with neutrons near the 14.2 MeV threshold of the ^{232}Th (n, 2nf) reaction .

Mono-energetic neutrons between 12.8 and 16.8 MeV were obtained with the CBNM Van de Graaff accelerator in the way as described by Liskien and Paulsen (22). Relative neutron fluences during the irradiation were monitored with a long counter.

The result of the angular dependence of the response of the dosimeter with one Th disc at 16.75 $\pm$ 0.20 MeV is given in figure 6. The measurements at 12.76 $\pm$ 0.14 MeV and at 14.24 $\pm$ 0.26 MeV show about the same shape, as indicated for the 14.24 MeV neutrons by the dotted line in this figure. A comparison of the angular response curve at 16.8 MeV and for ^{252}Cf neutrons shows that the response ratio 270° to 180° is somewhat larger for 16.8 MeV, whereas in the 16.8 MeV figure the response is peaked in the forward (270°) direction.

Measurements of the response ratio at 270° to 180° at different energies around 14 MeV are given in figure 7. The indicated error is the quadrature sum of the statistical

error, an uncertainty of 1.0 mm in the distance to the target and a 3% uncertainty in the monitor response. The results show a small increase in the 270° to 180° response with increasing neutron energy, reaching a maximum value between 15.0 and 15.5 MeV. The theoretical curve for the anisotropy of fission, indicated by the dotted line, has a much more pronounced peak near 14.2 MeV (20). This indicates that due to integration over a large angle of the fission products, the energy dependence of the angular anisotropy of the dosimeter response is not as large as that of the fission process itself.

Response ratios 270° to 90° from more accurate simultaneous irradiations of two thorium discs at several neutron energies, are also given in figure 7. The figure shows that there clearly exists a forward/backward effect at these neutron energies for our foil thickness and etching conditions, although the effect is much smaller than the ratios measured by Pauw (13).

3.3. Measurements with other fissile materials and mono-energetic neutrons between 12.8 and 16.8 MeV

The measurements of Pauw were carried out at 15.6 MeV with thin layers of ^{239}Pu, ^{237}Np and natural uranium. It was therefore decided to repeat these measurements at energies around 15 MeV with ^{237}Np and natural uranium with the same samples as used by Pauw. Also thin layers of enriched ^{235}U (99.501%) were measured. The fission layers were electroplated either on 1.0 mm thick nickel discs (Np and nat. U) or on 0.2 mm thick Al discs (^{235}U), and had a thickness of about 0.15 mg cm^{-2} and a diameter of about 13 mm. The amount of fissile material was determined with a maximum uncertainty of 2% by means of $2\pi\alpha$-counting at the CBNM. The fission track measurements were corrected

for the difference in mass and the difference in distance to the target.

The results of the simultaneous irradiation of the fission layers under 270° and 90° at 12.8, 14.2 and 16.8 MeV are given in figure 8. The statistical error was rather large as only a few hundred sparks were counted after irradiation times of 2-5 hours. If the forward backward effect is present, it is much smaller than the ratios measured by Pauw for the same nuclides.

The reason for the effect is not known. Recent measurements of Heinzelmann and Schüren (23), show an increasing ratio of 270° to 90° response with decreasing etching time, for a 12 μm thick Makrofol foil in contact with their Th-dosimeter and irradiated with AmBe neutrons. This would suggest that only the first few microns of the Makrofol foil are responsible for the effect, which might indicate that more heavy fission fragments are emitted under large angles in the backward than in the forward direction. In order to study this effect in a better way, irradiations with plastic foils placed at larger distances to the sample in a vacuum box would have to be carried out.

Measurements of the response ratio of 270° to 180° for ^{237}Np, ^{235}U and ^{238}U are also presented in figure 8. These results show that the anisotropy of the fission products measured by means of track registration in plastic foils in direct contact with the fissile layers, differs not very much for even-even or even-odd nuclides. The energy dependence seemed to be smaller for ^{237}Np and ^{235}U than for ^{238}U. More experiments will be necessary with better statistical accuracy e.g. by the use of thicker T targets in order to study the anisotropy of this type of dosimeter.

3.4. Directional dependence of the response of the final dosimeter

In the final dosimeter one Makrofol foil is sandwiched between two thorium discs. The forward/backward effect will thus disappear and the angular response will be symmetrical around 180°.

A spherical dosimeter would show no angular dependence of its response, but from a practical point of view such a type of dosimeter, or even a cube, which is a good approximation of a sphere, is difficult to realize. For the use of the dosimeter with ^{252}Cf sources, we therefore deduced from figure 5 and figure 6 a factor 1.25 ± 0.05 with which the sensitivity for perpendicular irradiation has to be multiplied in order to get the average response for irradiation under all angles.

4. Determination of the spectrum of scattered neutrons inside the glove box during the handling of ^{252}Cf sources

During the handling of ^{252}Cf sources the dose equivalent received will in general not only be due to neutrons coming directly from the source, but also from neutrons scattered in surrounding materials. Most of the neutrons in the Cf spectrum have energies of only a few MeV. After one or a few collisions the scattered neutrons may have an energy below the threshold of the Th(n, f) reaction (about 1.5 MeV), but they may still contribute to the total dose equivalent. This will especially be the case after collisions with hydrogen atoms. As the glove boxes have a water shield in order to minimize the neutron flux density outside the box, it seemed worthwhile to measure the neutron spectrum inside the box, in order to determine what fraction of the total dose equivalent is due to scattered neutrons below the threshold of the Th(n, f) reaction. This would give a factor with which the measured fast neutron dose equivalent should be multiplied in order to get the total dose equivalent received.

The spectrum of the scattered neutrons was determined by means of integral spectrum measurements. The ratio of the saturation activity measured inside the glove box to that in a low scattering environment for a detector at a certain distance from a ^{252}Cf source can be given by:

$$1 + \frac{\int \varphi(E)\ \sigma(E)\ dE}{\int \varphi'(E)\ \sigma(E)\ dE} ,$$

where $\varphi(E)$ and $\varphi'(E)$ are the flux densities of the scattered neutrons and the Cf neutrons respectively at neutron energy E, and $\sigma(E)$ is the detector cross section at that energy. If a set of detectors is used which have different treshold energies and well known excitation functions, than a scattered neutron spectrum $\varphi(E)$ versus E can be deduced from the measured ratios. As these measurement are relative, an efficiency calibration of the counting equipment is not needed.

Use was made of the $^{197}Au(n,\gamma)\ ^{198}Au$ and $^{115}In(n,\gamma)\ ^{116m}In$ reactions, which have a maximum in their energy response in the keV region, if the indium an gold detectors, which were massive spheres with diameter one cm and weight G, are surrounded by a $2.2.g.cm^{-2}$ thick ^{10}B shell. Also the $^{115}In(n,n')\ ^{115m}In$ reaction was used, which has a threshold around 500 keV. The excitation curves necessary for the calculations, were taken from the work of Pauw (13) and are given in figure 9.

The activations were performed at distances varying between 5 and 15 cm from a 10 μg ^{252}Cf source at a height of about 10 cm above the bottom of the glove box. The Cf source consisted of 10 μg Cf evaporated in a rather large stainless steel container with outer diameter 1.7 cm, height 7.0 cm and a wall thickness of 0.07 cm. In order to imi-

tate the real working conditions, also a paraffin hand was placed on both sides of the source at 5 cm distance from the detector. The gold and indium activities were measured in a 3 x 3 inch NaI(Tl) well-type crystal. The ^{116m}In was counted in the gamma -ray energy range from 700 to 2430 keV, whereas the ^{115m}In was counted, after a few hours, between 200 and 500 keV. The gold activity was measured in the gamma-ray energy range 292-518 keV. Accurate decay measurements showed a small fraction of ^{196m}Au activity, for which correction factors could be obtained from long irradiations, as described by Pauw.

After correction for the decay of the ^{252}Cf source during the measurements $\frac{Tr^2}{G}$ was calculated and plotted as a function of r. r is the distance between the centre of the source and the centre of the detector, and T is the saturation count rate. This curve shows a small increase with the distance for the (n, n') reaction both in the glove box and in the low scattering environment. This can be explained by assuming that the Cf is not in the middle of the container, but somewhat off centre. Taking into account the real distance r' to the place of the Cf, plots of $\frac{Tr'^2}{G}$ as a function of r' are given in figure 10. The In(n, n') measurements show that there are hardly any scattered neutrons above the threshold of this reaction. The ratio of the response glove box to low scattering environment is higher for gold than for indium. This is due to the fact that the Au(n,γ) + ^{10}B detector is more sensitive to scattered neutrons than the In(n, γ) + ^{10}B detector, because the indium response curve is more or less flat between 200 keV and 2 MeV, whereas the gold excitation curve rises rapidly with decreasing energy, as can be seen from figure 9.

Part of the extra count rate after activiation in the glove box may be due to neutrons scattered back from the one hand near the detector, and has to be subtracted from the total count rate in order to determine the real spectrum at the position of the detector. Therefore the ratio of the responses

with and without a paraffin hand at 5 cm from the gold or indium detectors, was measured at several distances, in the same way as described before. These measurements were performed using a small 1.4 μg ^{252}Cf source, a description of which is given elsewhere (24). The induced activities were therefore small, introducing a rather large error in the ratio. The results are given in table I.

Table I

Ratio of the response with and without one hand at 5 cm from the detector at different source detector distances.

	r = 4.0 cm	r = 8.0 cm	r = 12.0 cm
$^{115}In(n,\gamma)^{116m}In$	1.029 ± 0.010	1.065 ± 0.020	1.150 ± 0.046
$^{197}Au(n,\gamma)^{198}Au$	-	1.002 ± 0.025	1.027 ± 0.031

Contrary to the former measurements, here the ratio of the indium meausurements seemed to be somewhat higher than for gold, indicating that most neutrons that are scattered back from the hand have energies in the range where the ratio $\sigma_{In}(E)$ to $\langle\sigma\rangle^{Cf}_{In}$ is higher than $\sigma_{Au}(E)$ to $\langle\sigma\rangle^{Cf}_{Au}$ e.g. around 1 MeV.

In order to determine a spectrum from integral measurements with detectors having different thresholds, it is necessary to make assumptions concerning the shape of the spectrum to be expected, and to modify this spectrum in order to fit the measurements. As a first trial spectrum we used a 16 group Monte Carlo calculation, in which the irregular shape of the glove box was approximated by cylinders of different radii (25). The resulting spectrum of scattered neutrons consisted of 1/E - spectrum below about 10 keV, and a peak of higher energy neutrons around 800 keV, the height of which was dependent on the radius of the cylinder. One of these spectra was modified in order to find a spectrum relative to a ^{252}Cf neutron

spectrum which gave the best agreement for the response ratios glove box to low scattering environment for the three threshold reactions.

The ^{252}Cf spectrum used for these calculations was taken as $\varphi(E) = k.E^{1/2}.\exp(-E/1.41)$, for neutron energies above 1.5 MeV, whereas below 1.5 MeV the data of Zamyatmin et al. (26) were normalized to this Maxwellian distribution. For the average fission cross sections the data of Pauw (27) were used, which amounts for the $^{115}In(n,\gamma)$, $^{197}Au(n,\gamma)$ and $^{115}In(n,n')$ reaction with ^{10}B: 118.9 ± 4.5 mb, 83.1 ± 2.3 mb and 175 ± 7 mb respectively.

Table 2

Calculated and measured ratios of the response of the three threshold detectors at 10.0 cm from the ^{252}Cf source.

	$\left[\frac{\text{Glove box + 2 hands}}{\text{low scattering}}\right]_{exp.}$	$\left[\frac{\text{+ hand}}{\text{- hand}}\right]_{exp.}$
$^{115}In(n,\gamma)^{116m}In$	1.725 ± 0.042	0.094 ± 0.025
$^{197}Au(n,\gamma)^{198}Au$	1.971 ± 0.029	0.011 ± 0.029
$^{115}In(n,n')^{115m}In$	1.030 ± 0.014	-

	$\left[\frac{\text{Glove box + 1 hand}}{\text{low scattering}}\right]_{exp.}$	$\left[\frac{\text{glove box + 1 hand}}{\text{low scattering}}\right]_{calc.}$
$^{115}In(n,\gamma)^{116m}In$	1.631 ± 0.049	1.578
$^{197}Au(n,\gamma)^{198}Au$	1.960 ± 0.041	1.982
$^{115}In(n,n')^{115m}In$	1.030 ± 0.014	1.036

For the spectra plotted per unit lethargy as given in figure 11, the resulting ratios are given in table 2. Also at 5 and 15 cm from the ^{252}Cf source, the calculated ratios of the detector responses in the glove box to that in the low scattering environment agreed within the experimental error. This means that the spectrum of scattered neutrons does not vary greatly between 5 and 15 cm distance from the source.

As can be seen from figure 11, the scattered neutrons have energies below the Th(n, f) threshold and will thus not be detected by the dosimeter, as was confirmed experimentally. The resulting dose equivalent was calculated from this spectrum using the flux density to dose equivalent rate conversion factors as given in NBS Handbook 63 (1957), is given in table 3 as a fraction of the dose equivalent due to neutrons coming directly from the ^{252}Cf source. Also given in this table is the dose equivalent due to thermal neutrons, which was measured at 10 cm from the source by means of gold foils, and which was assumed to be constant in the box. The dose equivalent due to 1/E-neutrons at 10.0 cm was confirmed by cadmium covered gold foil measurements. In practice monitoring the dose equivalent due to thermal and 1/E-neutrons can thus be omitted, as they will contribute only a few percent at maximum to the total dose equivalent.

Table 3

Dose equivalent due to scattered neutrons.

	Relative to dose equivalent from unscattered neutrons				Absolute per μg ^{252}Cf
distance	thermal	0.5eV-10keV	10keV-max	total	$mrem.hr^{-1}$
5.0 cm	0.08 %	0.21 %	7.8 %	8.1%	72.0
10.0	0.30	0.64	23.6	24.5	54.3
15.0	0.68	1.23	45.0	46.9	46.2

The last column of table 3 gives the scattered dose equivalent in mrem.hr^{-1} for one μg of ^{252}Cf inside the box, using a flux density to dose equivalent rate conversion factor of 0.119 mrem.hr^{-1} per n.cm^{-2}sec^{-1}. As can be seen from these data, the extra dose equivalent due to scattered neutrons does not decrease very much with increasing distance from the source. This means that one cannot simply multiply the recorded fast neutron dose equivalent by a constant factor in order to get the total dose equivalent as can be done outside the box (3). Rather an average value of 50 mrem.hr^{-1} per μg ^{252}Cf inside the box, has to be added to the recorded fast neutron dose equivalent.

An estimate of this extra unrecorded dose equivalent received by the operator on his hands can be made if the total amount of ^{252}Cf inside the box is known and the time during which operations are carried out inside the box. It is evident that quantities of ^{252}Cf larger than 10 μg will give too high an unrecorded dose equivalent on the hands, and the use of manipulators will be necessary. If the californium is in a H_2O solution or otherwise shielded inside the box, then the quantity of 50 mrem.hr^{-1} per μg ^{252}Cf may change, but as the moderating layers in general will not be very thick, large variations are not expected, though experimental confirmation will be necessary.

The same is valid in the case of the handling of ^{252}Cf needles for therapy purposes. However here an experimental determination of the total dose equivalent at the position of the hands under working conditions may be possible by means of a remcounter. This may give after subtraction of the direct ^{252}Cf dose equivalent, the dose equivalent due to scattered neutrons.

Though the threshold of the ^{237}Np(n, f) reaction is lower than that of the ^{232}Th(n, f) reaction, about 0.5 MeV for Np compared to 1.5 MeV for Th, the In(n, n') measurements show

that the scattered neutrons will not be recorded by a Np-dosimeter either e.g. the one proposed by Sohrabi and Becker(28). The $^{103}Rh(n,n')^{103m}Rh$ reaction has a lower theoretical threshold: about 50 keV. However, due to its low sensitivity and the rather short half-life of the ^{103m}Rh (56 min.) a dosimeter based on this reaction is only suitable for special purposes (29), e.g. the opening of ^{252}Cf-capsules. Although albedo type dosimeters are inferior to fast neutron dosimeters for measuring the neutron dose equivalent on the body of the operator outside thin water shields (3), they may be used together with a fission track dosimeter on the hands inside a glove box for measuring the extra dose equivalent due to scattered neutrons. However, an experimental determination of the sensitivity inside the box will be necessary, whereas also other factors like the influence of the thickness of the hand on the response has to be investigated.

The spectrum of the scattered neutrons and its important contribution to the total dose equivalent, emphasizes once more the lack of a personnel neutron dosimeter system having a threshold near about 10 keV.

5. Sensitivity of the dosimeter.

The response of the dosimeter in a pure ^{252}Cf spectrum was determined by irradiating the dosimeter perpendicular to the thorium discs in a low scattering environment at different distances from a small, calibrated ^{252}Cf source which will be described elsewhere (24). The resulting sensitivity, which is of course only valid for our dimensions of the thorium discs, foil thickness, etching and counting conditions, amounts to $(2.78 \pm 0.06) \times 10^{-6}$ sparks.cm^2.n^{-1}, which is $(0.885 \pm 0.019) \times 10^{-6}$ sparks.cm^2.n^{-1} for a Th disc of one cm^2.

In order to convert the counted number of sparks to dose equivalent, which is the quantity of interest from the point of view of radiation protection, Prêtre (8) prefers to

calibrate his dosimeter in rad using the maximum absorbed dose to fluence conversion factor. Afterwards the measured dose can then be converted into dose equivalent using appropriate quality factors. Buys et al. (9) gave an experimentally determined conversion factor holes. (foil. rem)$^{-1}$ using an average number of counted holes in foils irradiated under different experimental circumstances in which the dose equivalent was measured by means of a remcounter. In our case, in which only the fast neutron dose equivalent due to neutrons coming directly from the Cf-source is measured, an average dose equivalent rate per unit flux density for ^{252}Cf neutrons of 0.119 mrem.hr^{-1} per n.cm^{-2}.sec^{-1} was calculated for the californium spectrum (section 4). This value is in good agreement with 0.122 mrem.hr^{-1} per n.cm^{-2}.sec^{-1}, as given by Stoddard and Hootman (30). If the fluence to dose equivalent conversion factors for monoenergetic neutrons for whole body neutron exposure, from which this factor has been calculated, will change [e.g. according to the proposal of Stevenson et al. (31)], a new conversion factor can be calculated for the known ^{252}Cf-neutron spectrum.

The resulting sensitivity will now be: 11.9 $\pm$ 0.3 mrem.spark^{-1} for ^{252}Cf neutrons impinging perpendicular on our dosimeter, which amounts to 37.4 $\pm$ 0.9 mrem.spark^{-1} for a Th disc of 1 cm^2. If one takes into account the factor of 1.25 $\pm$ 0.05 due to the angular dependence of the response as described in section 3, the resulting sensitivity will be: 14.9 $\pm$ 0.7 mrem.spark^{-1}, which has been used for our dosimeter readings. For a thorium disc of one cm^2 the sensitivity will be 46.8 $\pm$ 2.2 mrem.spark^{-1}. This value may be compared with the value of 91 mrem.spark^{-1} per cm^2 thorium for Cf neutrons for the dosimeter of Buys et al. and a sensitivity of 12.6 mrad.spark^{-1} per cm^2 thorium of the dosimeter

of Prêtre, which corresponds to a response of 107 mrem. $spark^{-1}$ using a quality factor of 8.5 for ^{252}Cf neutrons. Both dosimeters contain, however, only one thorium disc.

In order to obtain the total neutron dose equivalent, one has to add the dose equivalent due to scattered neutrons. As discussed in section 4 an estimated value of 50 $mrem.hr^{-1}$ per μg ^{252}Cf inside the box could be calculated. This extra contribution was not very dependent on the distance to the source.

Measurements with the ^{252}Cf source and the dosimeter combined with a TLD chip, provided the ratio of γ exposure to counted number of sparks, and amounted to 0.640 ± 0.023 $mR.spark^{-1}$, using a small correction factor for the somewhat different position of the TLD disc. This means that in air the measured ratio of the gamma dose equivalent to the neutron dose equivalent amounted to: $(4.67 \pm 0.20) \times 10^{-2}$ using a conversion factor of 0.869 for the γ-exposure and a neutron sensitivity of 11.9 ± 0.3 $mrem.spark^{-1}$. This is somewhat lower than the number of 7.03 % as given in (31) , which may be due to absorption of low energetic X and gamma rays in the encapsulation material of the neutron source, and in the 0.5 mm thick stainless steel container of the TLD chip.

Measurements of the dosimeter response with and without a paraffin dummy hand between the dosimeter and the Cf-source, showed that the neutron dose equivalent is lowered by a factor of about two by placing the dosimeter on the back of the hand. The measured gamma dose is somewhat higher in the situation with the dummy hand. This means that wearing a dosimeter on a hand will hardly influence the measurement of the gamma dose equivalent, whereas the measurement of the neutron dose equivalent will be different for the front and the back of the hand. However, as pointed out in the introduction, the dosimeter gives only a measurement of the neutron fluence at the place of the dosimeter whereas the choi-

ce of an appropriate dose equivalent conversion factor may give a dose equivalent averaged either over the whole hand or only at the position of the dosimeter (see for example (31)).

6. Experience with the dosimeter.

As at CBNM only a few people are working in glove boxes with Cf sources, not many results of recorded γ and neutron doses are available. Nevertheless some conclusions can already be drawn from the experience with the use of the dosimeter over a period of about one year.

Figure 12 gives the ratios of the measured TLD doses on the finger tip to that on the hand, both of which were corrected for background and radiation due to the thorium, for the left and right hand. As can be seen from this figure, our starting point was indeed correct, that in most cases the γ-dose on the hand is of the same order of magnitude or even higher than the γ-dose equivalent on the finger tips. Most cases where the γ-dose on the finger tips was higher than on the hand, occurred at special occasions in which the dosimeters were worn only during short periods of time e.g. the opening of a container with a quantity of 10 μg ^{252}Cf. Though the ratio of the neutron dose equivalent on the hands to that on the finger tips will differ from the gamma dose equivalent ratio (see below) it seemed nevertheless useful to wear a larger neutron dosimeter on the hand, in combination with TLD's at the finger tips and in the dosimeter.

Figure 13 gives the ratios of neutron dose equivalent to γ-dose equivalent measured on the back of the hand, using the neutron sensitivity as given in section 5. Though the statistical error in the neutron dose equivalent measurements was rather large, the maximum number of counted sparks being 60, the measured ratios showed a much larger spread. This may be due to differences in orientation of the hands with the dosime-

ter towards the source (cf. section 5). Also the ratio of the neutron fluence to the gamma fluence will in general be a function of the position in the box. This means that estimating the neutron dose equivalent by a simple measurement of the gamma dose equivalent on the hand with a TLD chip and multiplying the result with a certain factor, will give large deviations from the real neutron dose equivalent.

According to figure 13 the measured γ-dose equivalent on the hands is in most cases of the same ordes of magnitude or higher than the measured neutron dose equivalent. From the calibration measurements in a low scattering environment with a ^{252}Cf source as described in section 5, it followed that the direct γ-dose equivalent is only about 5% of the neutron dose equivalent. The largest part of the γ-dose equivalent will thus be due to scattered gamma rays or gamma's coming from absorption of thermal neutrons. Reduction of the gamma dose equivalent due to gamma rays from capture of thermal neutrons by hydrogen in the water shield, can be obtained by adding boron to the water. According to Stoddard and Hootman (30), a reduction to 40 % of the gamma dose outside a 15 cm thick water shield can be obtained by adding 2 mg ^{10}B per cm^3 H_2O, which is equivalent to about a saturated solution of H_3BO_3 (5.2 g H_3BO_3 per 100 ml H_2O at 21 °C). Experimental determination of the reduction of the gamma dose inside the box will, however, be necessary.

7. Conclusions

A personnel neutron dosimeter using fission track registration as described in this paper is not an ideal instrument, but at this moment it seems to be the best type of dosimeter for the measurement of the fast neutron dose equivalent on hands of operators during the handling of ^{252}Cf sources.

By a careful construction of the dosimeter and the coun-

ter thus avoiding touching the 10 μm thick Makrofol foil during the preparation of the dosimeter, by carefully selecting the etching and counting process, and by testing the foils before use, a zero background could be ontained. The sensitivity of the dosimeter for ^{252}Cf neutrons unter our etching and counting conditions amounts to 14.9 ± 0.7 mrem.spark^{-1}, taking into account a factor of 1.25 ± 0.05 for the angular dependence of the response of the dosimeter.

Though the anisotropy of fast neutron induced fission of ^{232}Th is dependent on the neutron energy and can be quite large near the onset of the (n, n'f) , (n, 2nf) etc. reactions, no large energy dependence of the angular response was found, due to the integrating effect of placing the plastic foil near the thorium disc .

A difference in response of about 20 % has been observed between irradiating a dosimeter with one thorium disc either with the Makrofol foil or the thorium disc towards the neutron source, at neutron energies between 12.8 and 16.8 MeV. For ^{252}Cf neutrons this forward/backward effect could not be observed.

Measurements of the spectrum of scattered neutrons inside the box shows that most scattered neutrons have energies between 10 keV and 1 MeV, and will thus not be recorded with a thorium (or neptunium) dosimeter.

The dose equivalent due to scattered neutrons amounts to about 50 mrem.hr^{-1} per μg ^{252}Cf inside the box, and is not very dependent on the distance from the source. It is therefore advisable to handle quantities of ^{252}Cf smaller than 10 μg in glove boxes, and to use manipulators for larger quantities.

The ratios of the measured γ-dose equivalent at the back of the hand and the γ-dose equivalent at the finger tip shows that in most cases the γ-dose on the hand is of the same order of magnitude as on the finger tip. It seems there-

fore justified to use a neutron dosimeter worn on the hand for use inside a glove box, which can be made larger (thus having a higher sensitivity) and which is more convenient than a neutron dosimeter attached to a finger.

The ratio of the measured neutron dose equivalent to the measured γ-dose equivalent at the back of the hand shows large variations. A simple calculation of the neutron dose equivalent received by multiplying the measured γ-dose equivalent with a certain factor is not satisfactory for work inside a glove box and a measurement of the neutron dose equivalent will be necessary.

In most cases the γ-dose equivalent measured inside the box is higher than the measured fast neutron dose equivalent. Reduction of the γ-dose equivalent due to gamma-rays from the capture of thermal neutrons by hydrogen, can be obtained by adding boron to the water shield.

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FIGURE CAPTIONS

Fig. 1 Exploded view of the CBNM hand neutron dosimeter

Fig. 2 CBNM dosimeter worn on a paraffin dummy hand

Fig. 3 ETC and frame with 10 μm thick Makrofol foil and aluminized Mylar foil

Fig. 4 Diagram of the electronic counting system of the ETC

Fig. 5 Angular dependence of the response of the dosimeter with one thorium disc for ^{252}Cf neutrons

Fig. 6 Angular dependence of the response of the dosimeter with one thorium disc for 16.75 MeV neutrons

Fig. 7 Response ratios 270° to 180° and 270° to 90° for dosimeters with one thorium disc at different neutron energies

Fig. 8 Response ratios 270° to 180° and 270° to 90° for dosimeters in contact with thin layers of ^{235}U, ^{237}Np and ^{238}U at three neutron energies

Fig. 9 Excitation functions of the three threshold reactions used for the determination of the spectrum of scattered neutrons

Fig. 10 $\frac{Tr'^2}{G}$ as a function of r' for the three reaction products inside the glove box and in a low scattering environment

Fig. 11 Neutron spectrum inside the glove box at 10.0 cm from a ^{252}Cf source

Fig. 12 Ratios of the gamma dose on the finger tip to that on the hand inside the glove box

Fig. 13 Ratios of the measured fast neutron dose equivalent to gamma dose equivalent on the hand inside the glove box

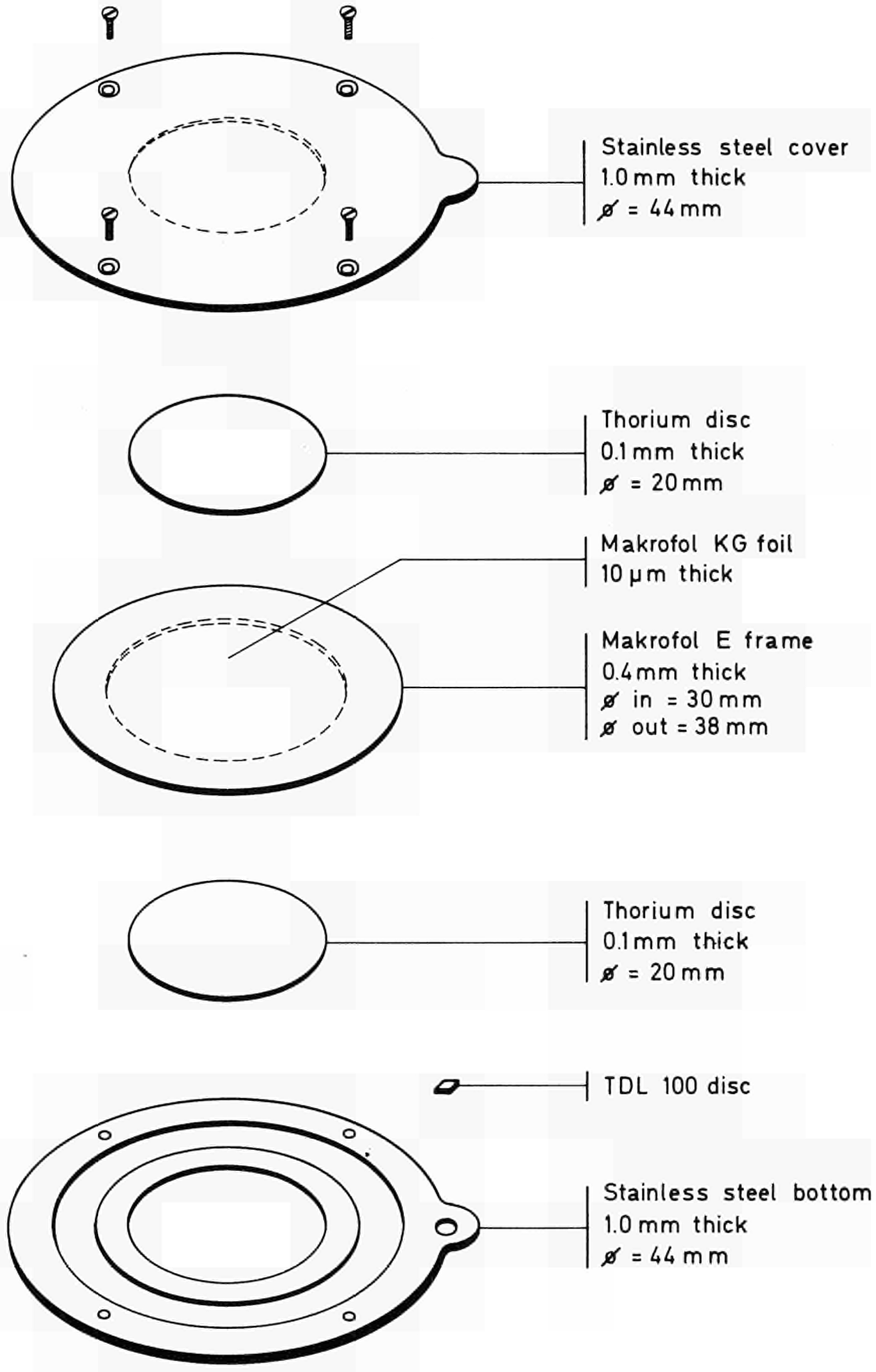


Fig. 1 Exploded view of the CBNM hand dosimeter

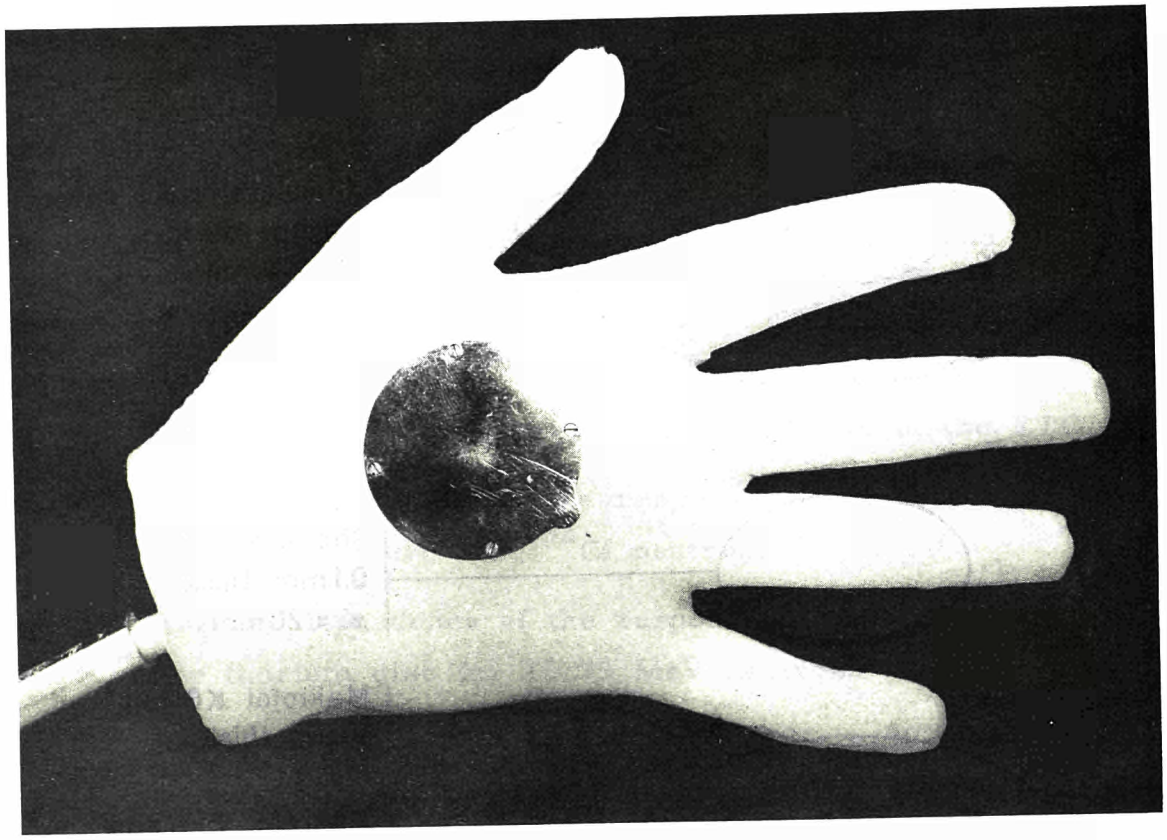


Fig. 2 CBNM dosimeter worn on a paraffin dummy hand.

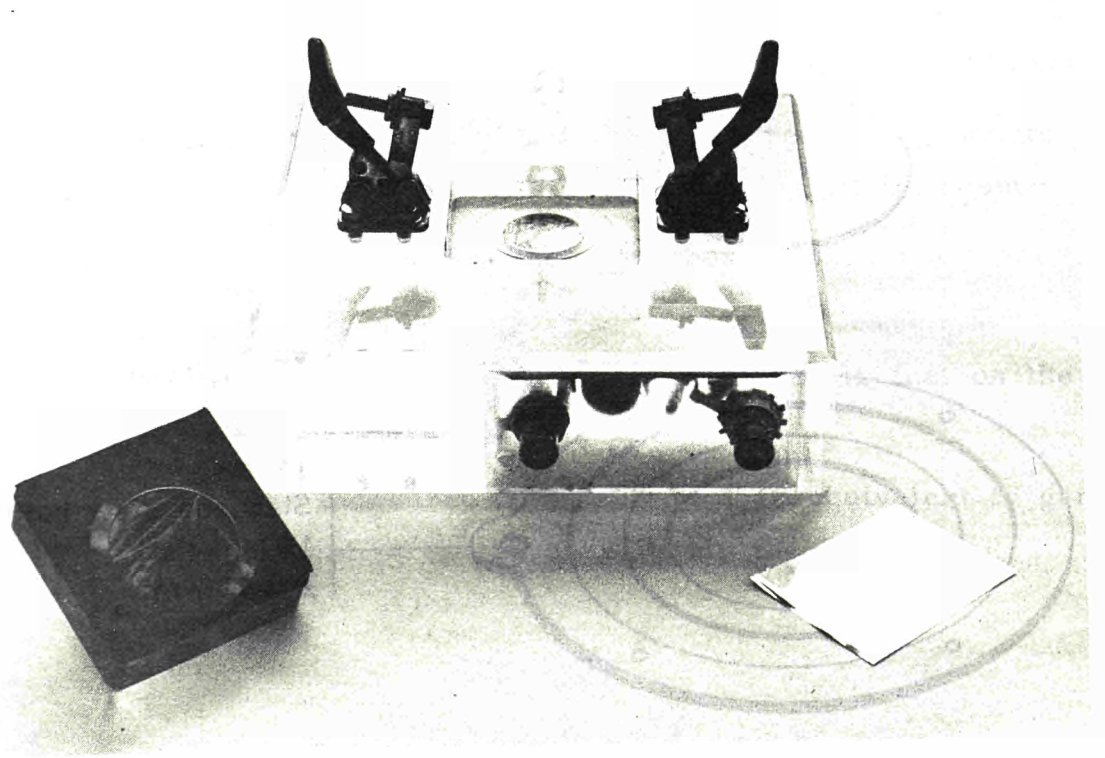


Fig. 3 ETC, frame with 10 µm thick Makrofol foil (left) and aluminized Mylar electrode (right).

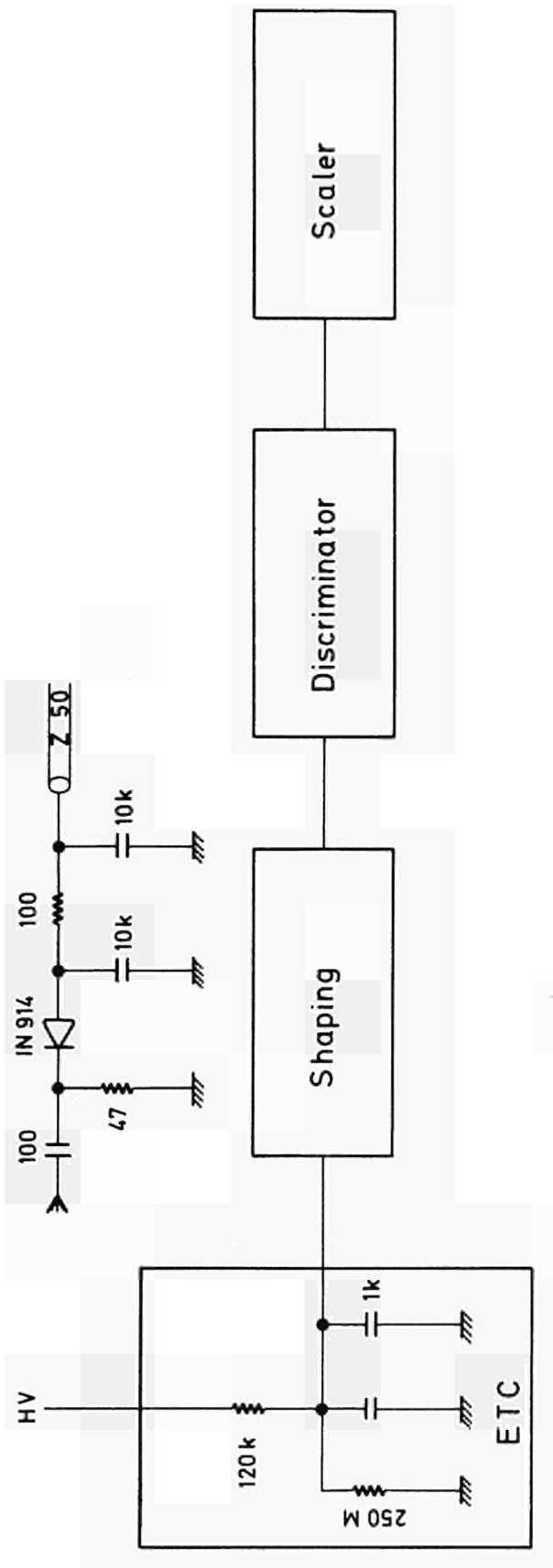


Fig. 4 Diagram of the electronic counting system

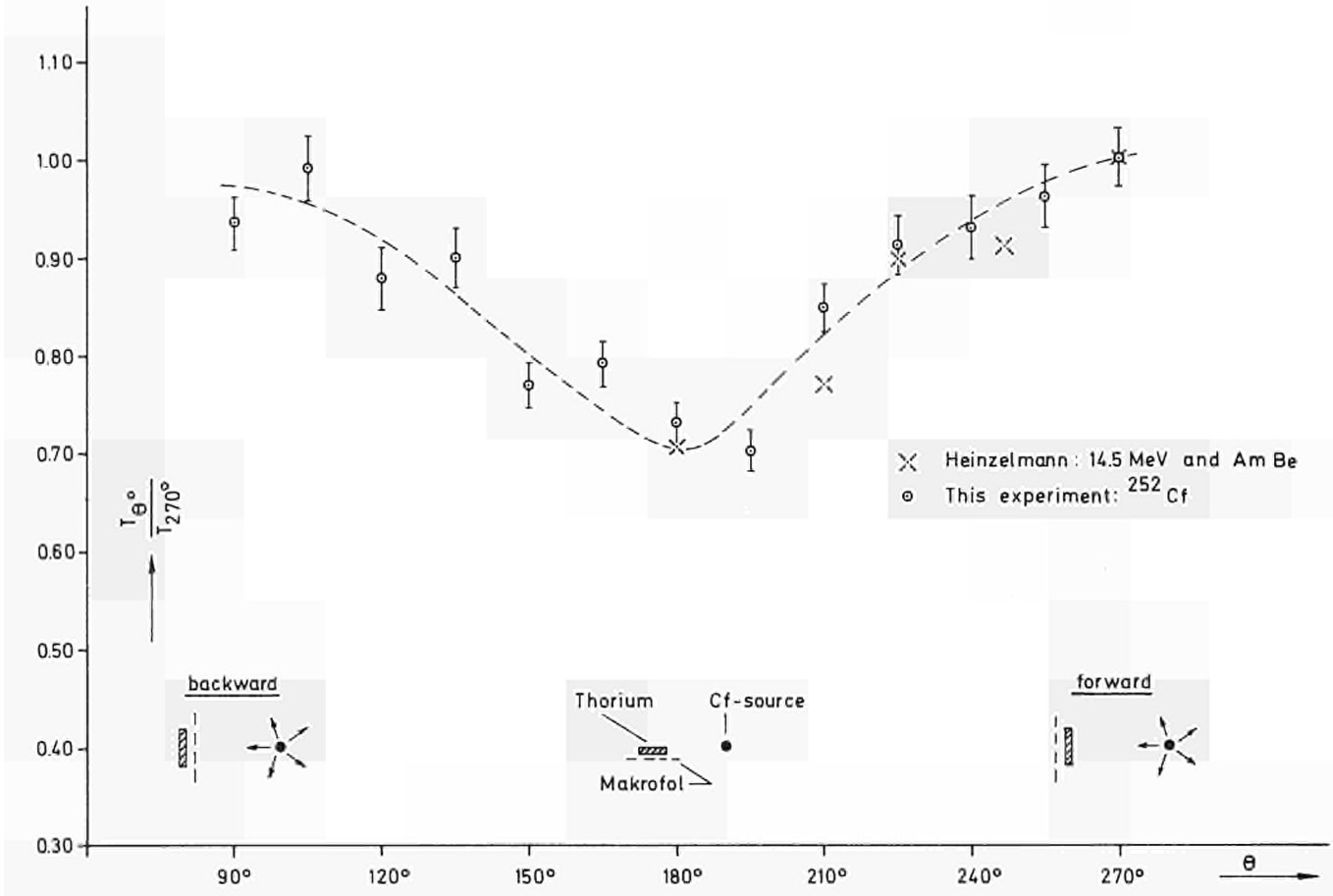


Fig. 5 Angular dependence of the response of the dosimeter with one thorium disc for ^{252}Cf neutrons.

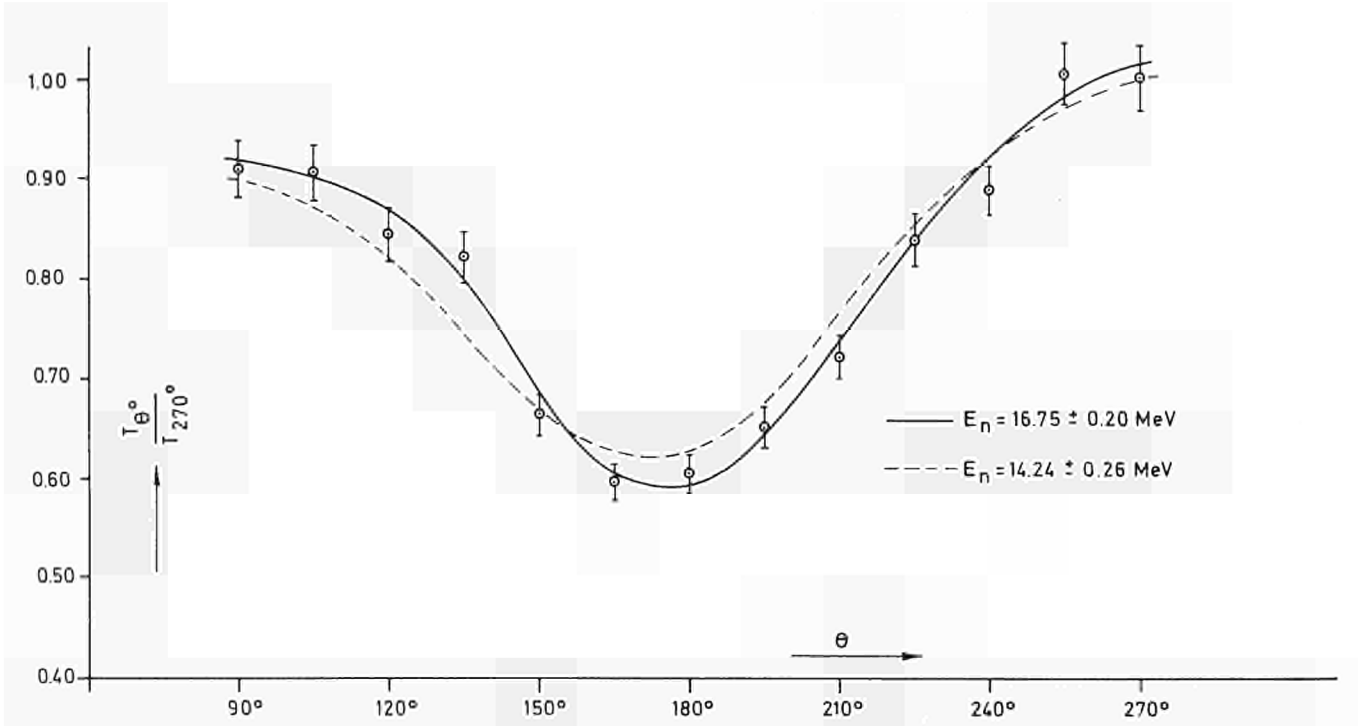
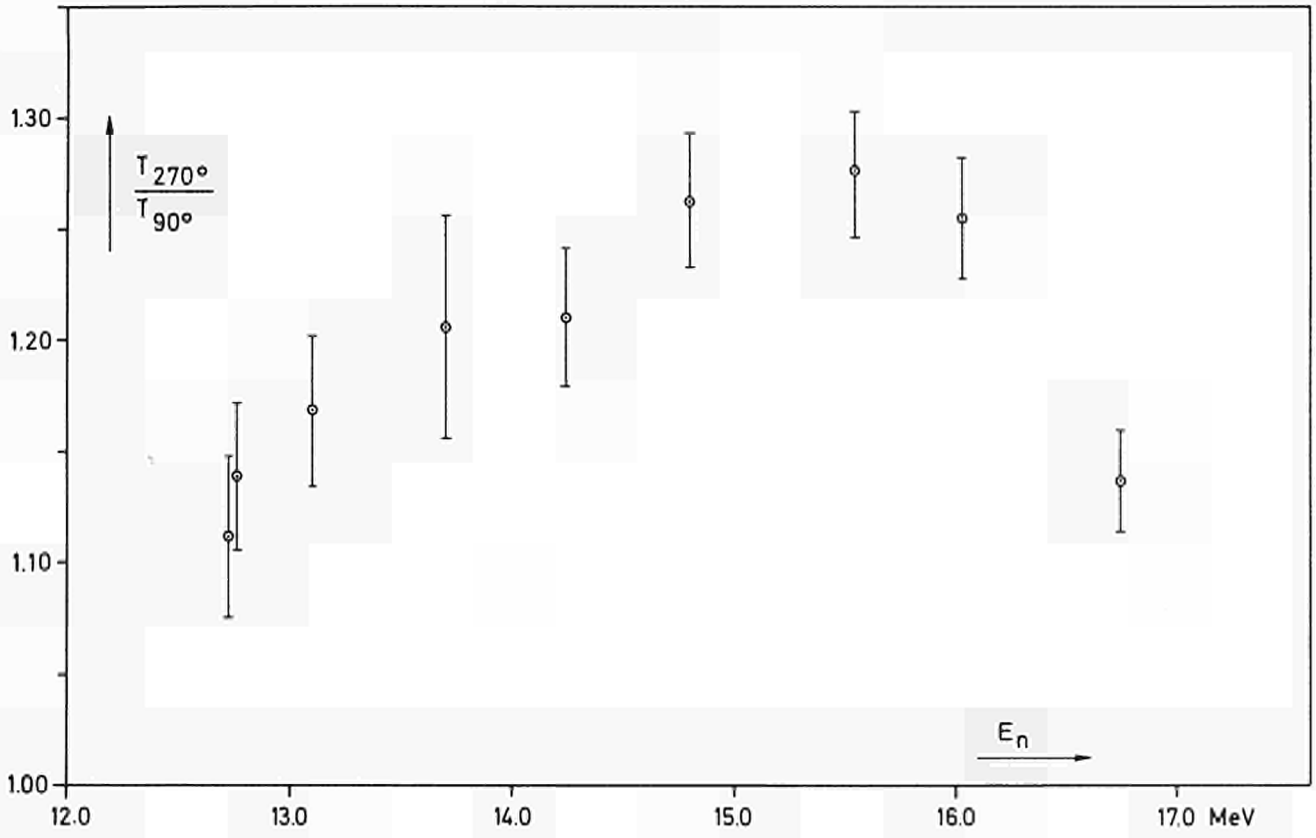


Fig. 6 Angular dependence of the response of the dosimeter with one thorium disc for 16.75 MeV neutrons.



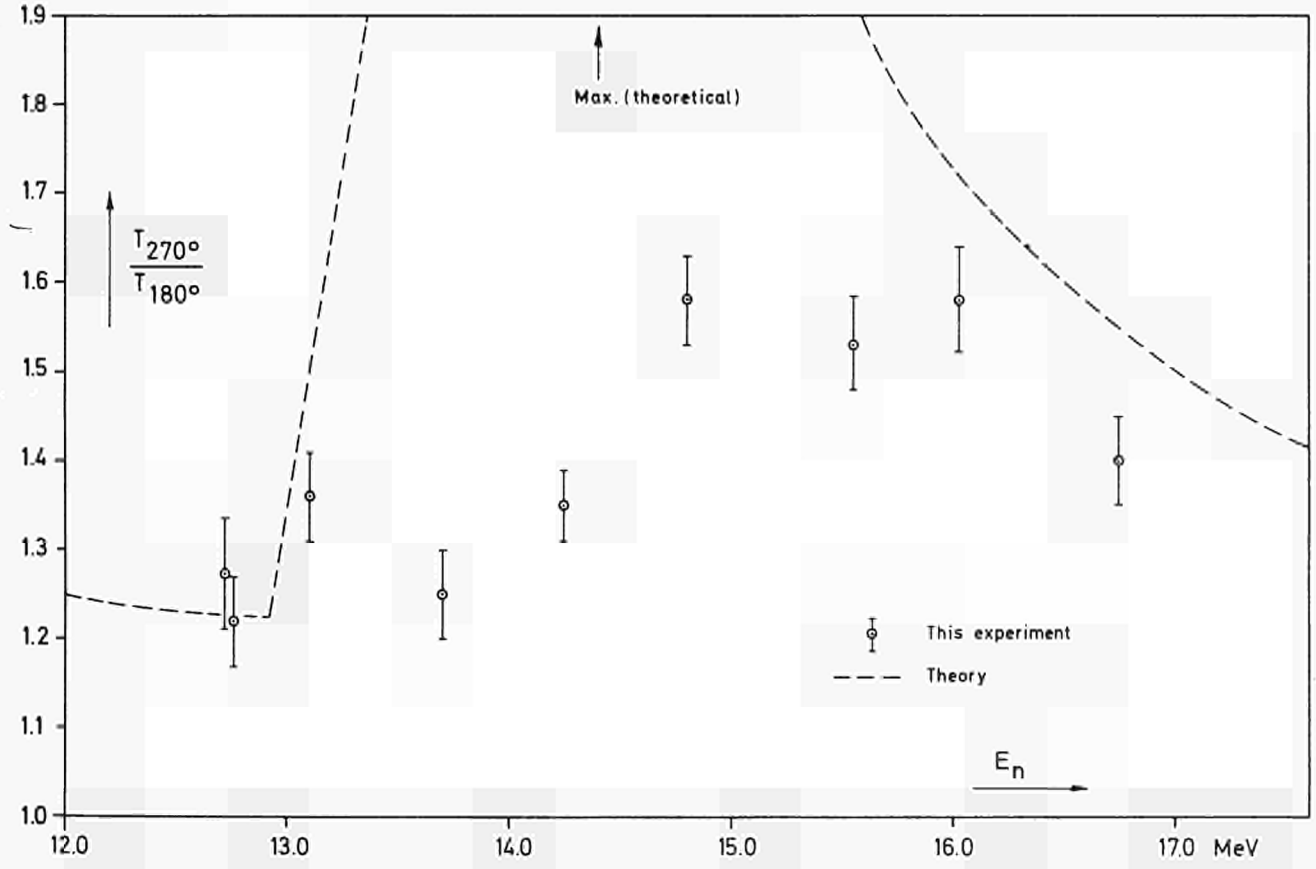


Fig. 7 Response ratios 270° to 180° and 270° to 90° for dosimeters with one thorium disc at different neutron energies.

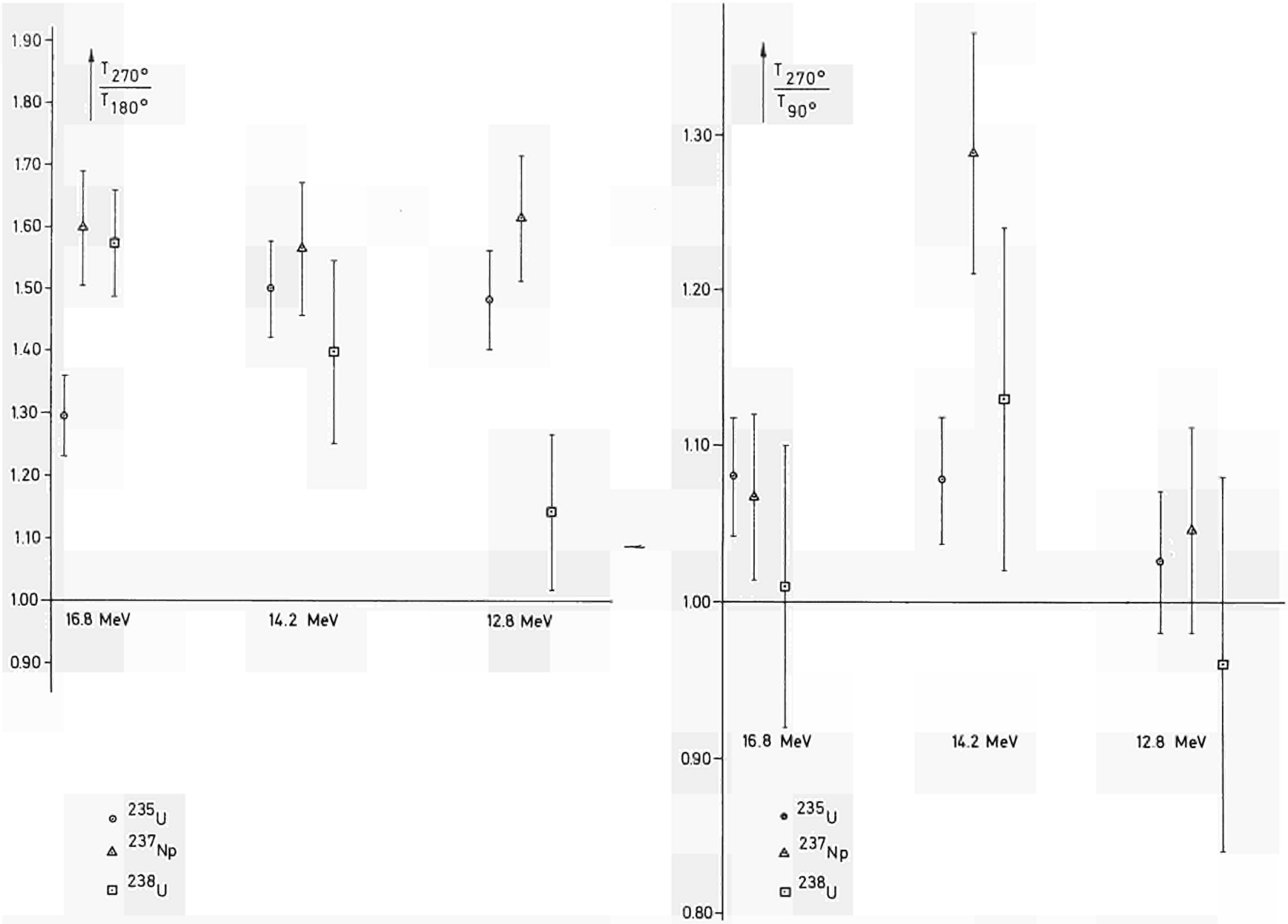


Fig. 8 Response ratios 270° to 180° and 270° to 90° for dosimeters in contact with thin layers of ^{235}U, ^{237}Np and ^{238}U at three neutron energies.

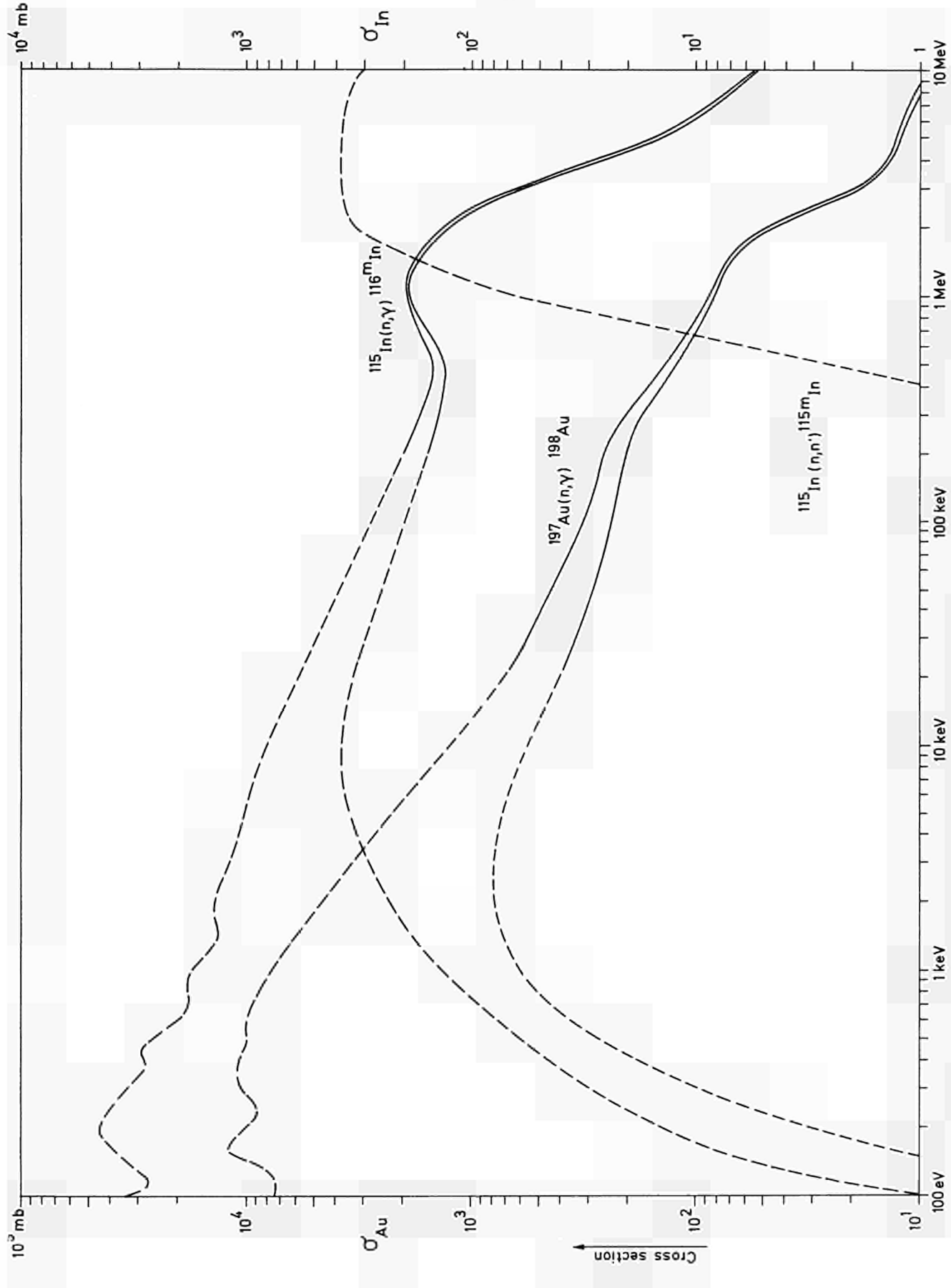


Fig. 9 Excitation functions of the three threshold reactions used for the determination of the spectrum of scattered neutrons.

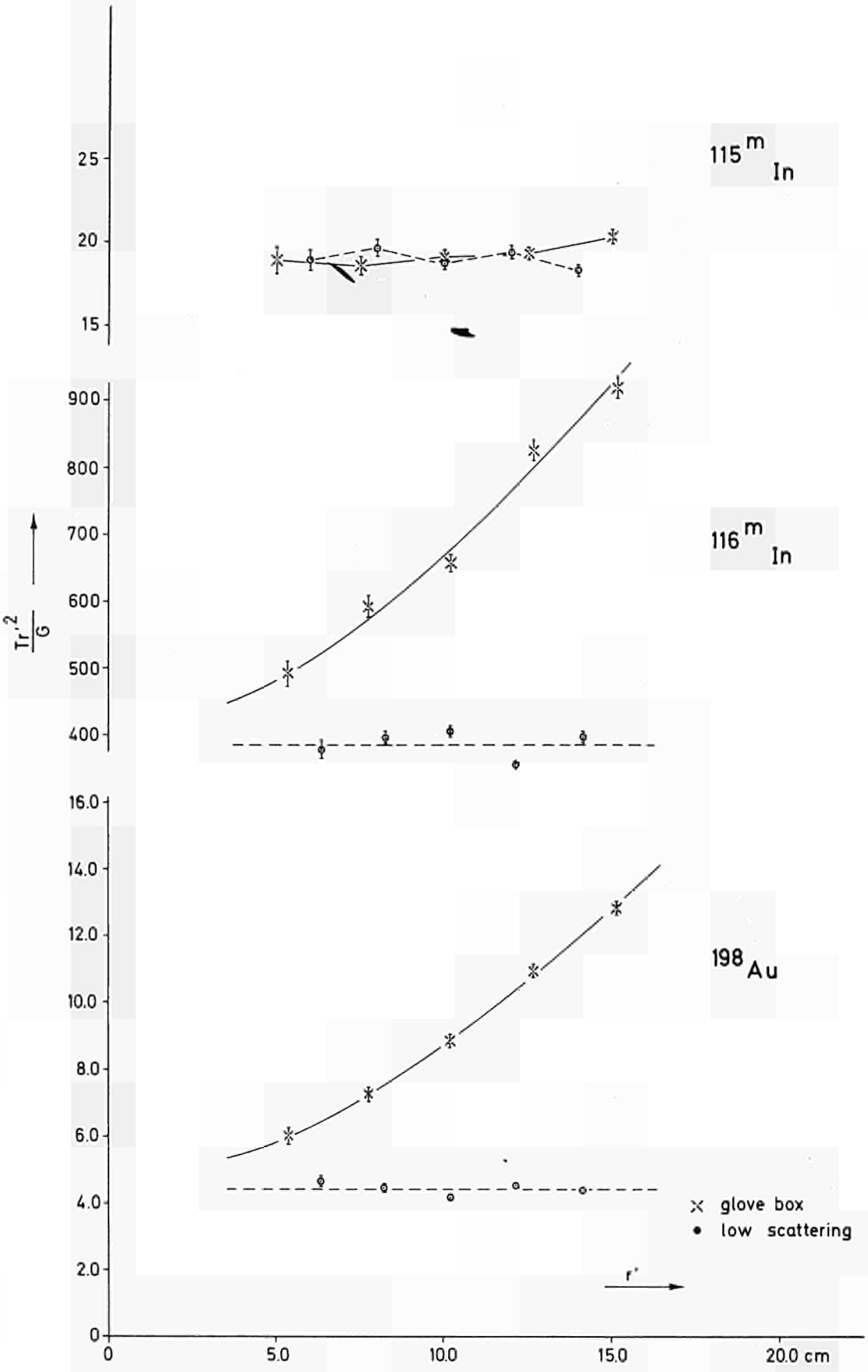


Fig. 10 $\frac{Tr'^2}{G}$ as a function of r' for the three reaction products inside the glove box and in a low scattering environment.

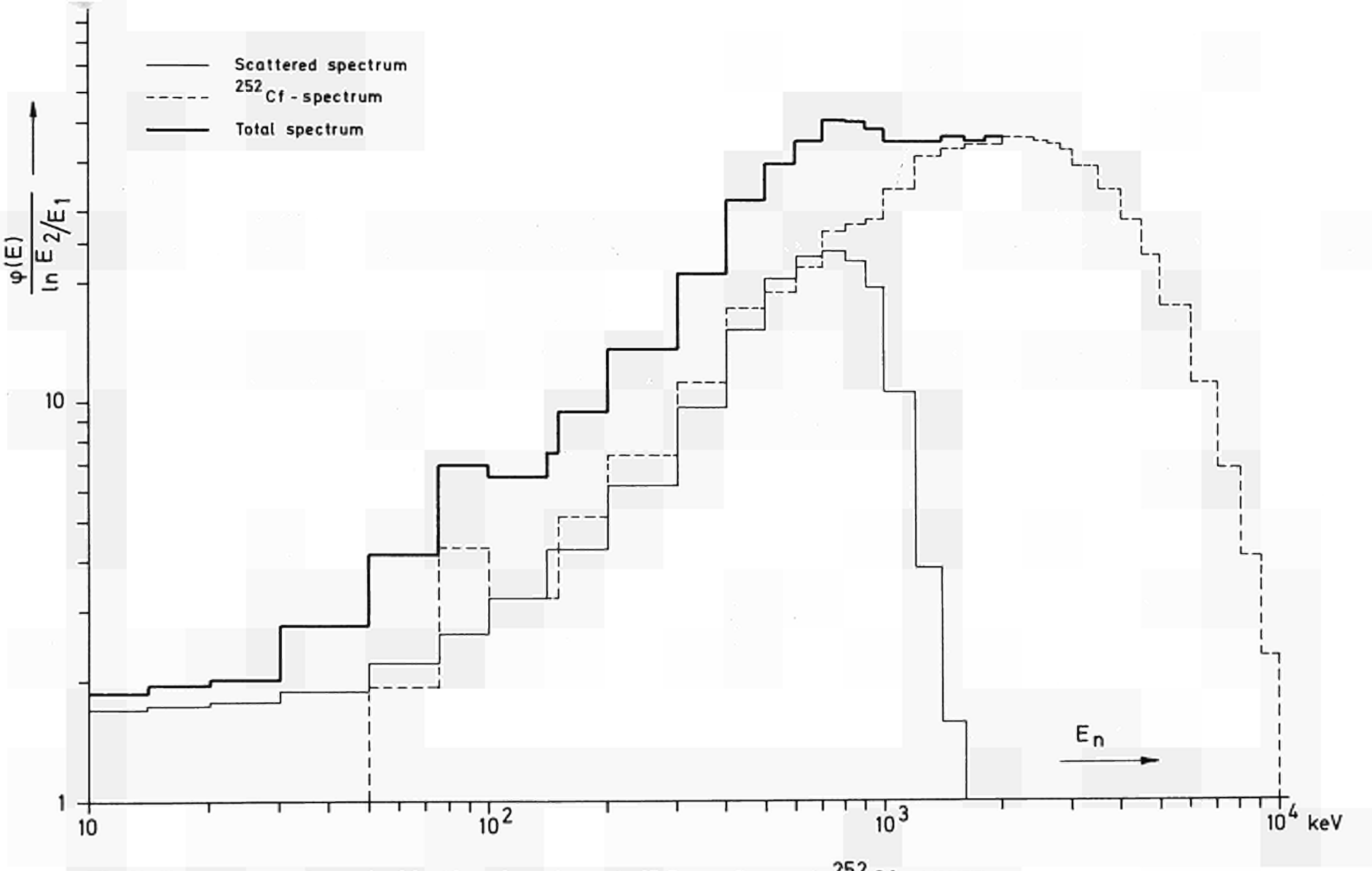


Fig. 11 Neutron spectrum inside the glove box at 10.0 cm from a ^{252}Cf source.

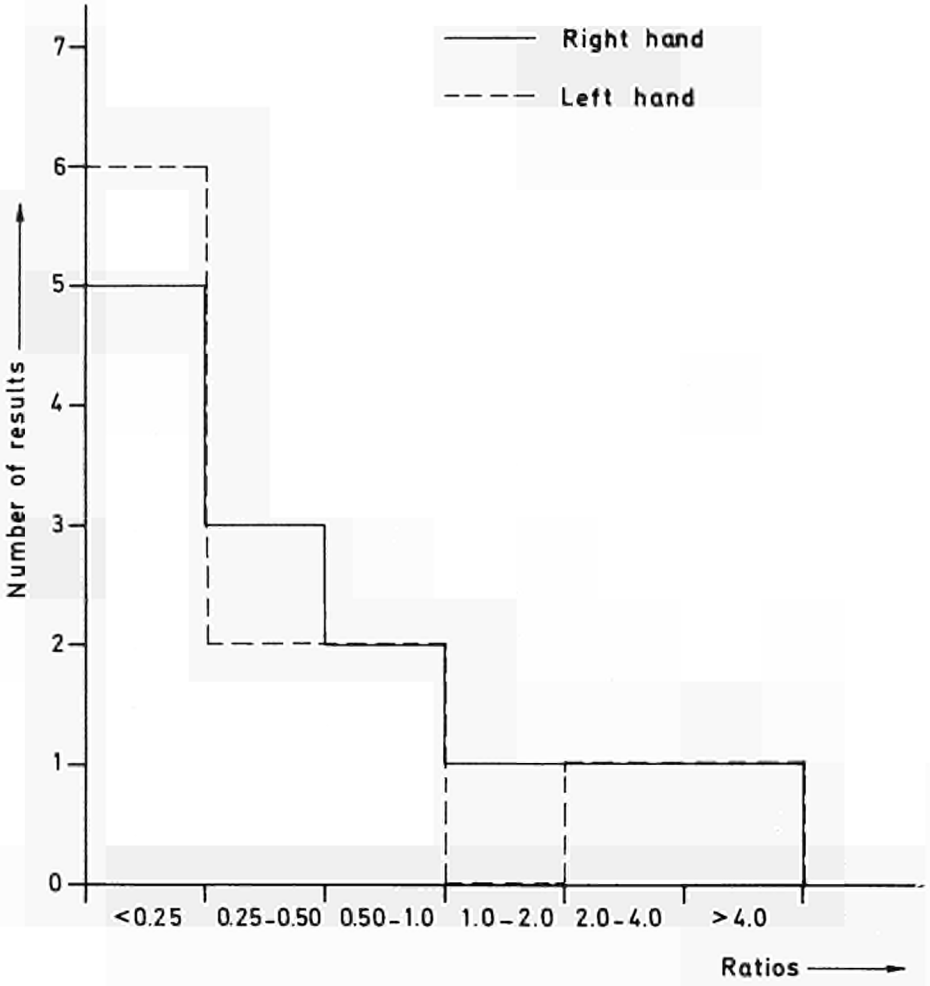


Fig.13 Ratios of the measured fast neutron dose equivalent to gamma dose equivalent on the hand inside the glove box

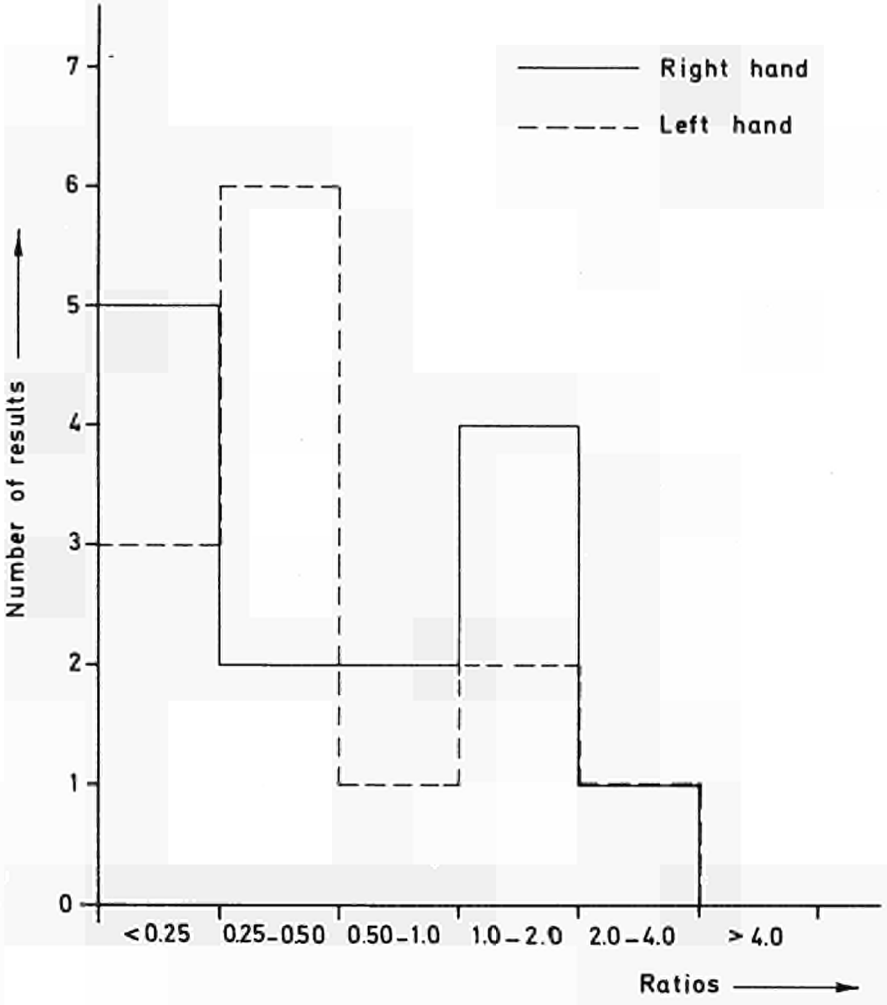


Fig.12 Ratios of the gamma dose on the finger-tip to that on the hand inside the glove box.

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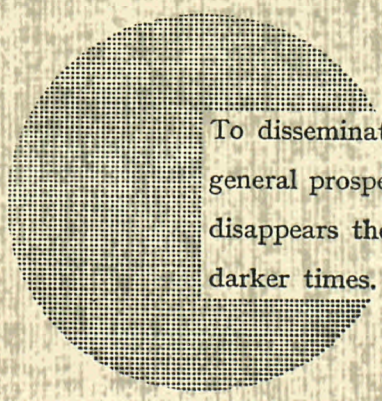

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Alfred Nobel

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