KERNFORSCHUNGSZENTRUM

KARLSRUHE

November 1970

KFK 1502

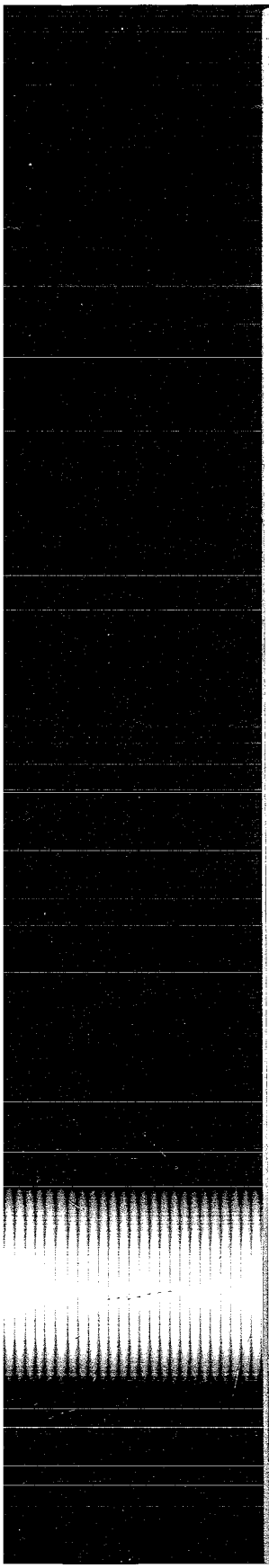
Abteilung Strahlenschutz und Sicherheit

Development of New Neutron Detectors for Accident Dosimetry

E. Piesch

GESELLSCHAFT FÜR KERNFORSCHUNG M.B.H.

KARLSRUHE



Reprint from

"ADVANCES
IN PHYSICAL AND BIOLOGICAL
RADIATION DETECTORS"

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA, 1971

DEVELOPMENT OF NEW NEUTRON DETECTORS FOR ACCIDENT DOSIMETRY

E. PIESCH
Gesellschaft für Kernforschung,
Kernforschungszentrum Karlsruhe,
Federal Republic of Germany

Abstract

DEVELOPMENT OF NEW NEUTRON DETECTORS FOR ACCIDENT DOSIMETRY.

New detectors and measuring techniques are proposed to improve the assessment of individual dose received from persons involved in a criticality accident. The aim was to reduce the number of detectors in the conventional detector combinations, to use sensitive activation reactions and to measure the dose of intermediate and fast neutrons directly. The proposed neutron detectors for the dosimeter combination are: (a) Arsenic, to detect slow and intermediate neutrons up to 1 MeV by the $^{75}As(n,\gamma)^{76}As$ reaction (half-life of ^{76}As is 26.4 h); (b) Phosphorus, to detect fast neutrons above a threshold of 2 MeV by the $^{31}P(n,p)^{31}Si$ reaction (half-life of ^{31}Si is 2.6 h), and slow neutrons by the $^{31}P(n,\gamma)^{32}P$ reaction (half-life of ^{32}P is 14 d); (d) A polycarbonate detector (Makrofol E) as a nuclear track detector to detect fast neutrons above a threshold of 0.5 MeV by elastic scattering and (n,α) reactions in carbon and oxygen. The β-activity of ^{76}As, ^{31}Si and ^{32}P can be measured directly in As_2S_3 glass and in phosphate glass by means of the β-induced Cerenkov effect. It uses a liquid scintillation counter set up as for tritium measurements. The calibration of the detectors was performed by calculations of the detector sensitivity for different neutron spectra and by irradiation with different neutron sources at different criticality installations. After an accident a first estimation of the neutron dose is obtained by a β-counting of the arsenic phosphate glass, which indicates the surface adsorbed dose or the total neutron fluence directly. It is energy independent over the range of intermediate and fast neutrons.

1. INTRODUCTION

As a rule, a criticality dosimeter system consists of a combination of activation detectors which allow evaluation of the neutron fluence over specific ranges of energy. The activation detectors are calibrated directly to suit an accident spectrum which, in the simplest case, is composed of three parts:

(i) An unmoderated Maxwell fission spectrum in the fast neutron range;
(ii) A 1/E spectrum in the range of intermediate neutrons from 1 MeV to 0.12 eV;
(iii) A thermal neutron spectrum with a Maxwellian energy distribution and a mean energy of 0.025 eV.

However, in a criticality accident the neutron spectrum at the place where a person is exposed is very markedly determined by:

(a) The type of critical assembly: its composition, thickness, and geometrical configuration of fuel elements, nature, and enrichment factor of fissile material;
(b) The type of moderation; metallic configuration of fissile material, fissile material in aqueous solution, fuel elements in the moderator;

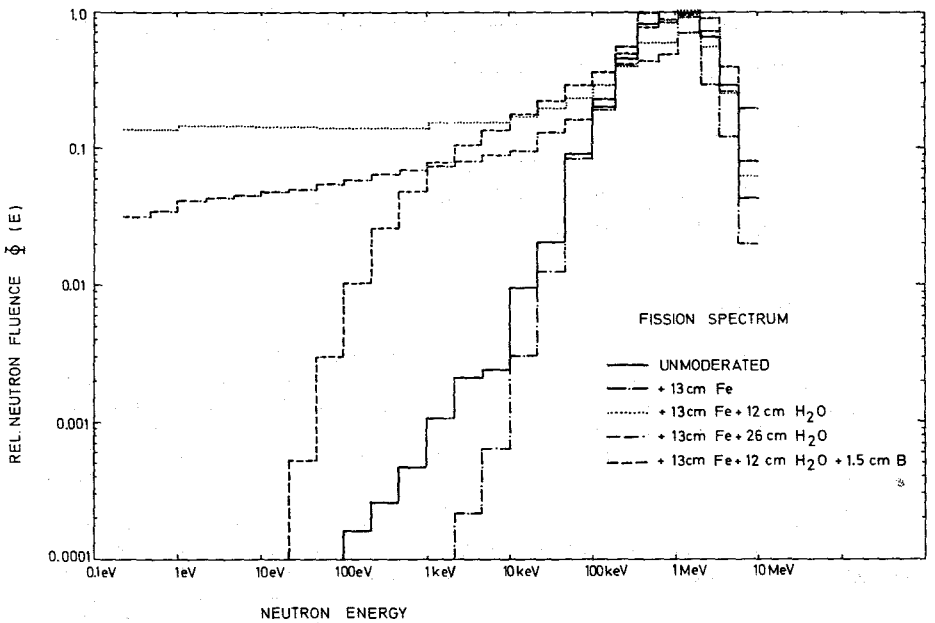


FIG. 1. Calculated leakage neutron spectra from an unmoderated critical system behind different shieldings [4].

(c) The type and thickness of additional shieldings: concrete, iron, aluminium placed between the critical assembly and the position of the exposed person.

A combination of threshold detectors is used to determine the neutron spectrum. The "Threshold Detector Unit" (TDU) of Oak Ridge [1, 2] is used as a free-air reference dosimeter. It consists of a combination of fissile foils of ^{239}Pu, ^{237}Np, ^{238}U and ^{32}S contained in a boron capsule. This provides energy thresholds of 0.5 keV, 0.75 MeV, 1.5 MeV and 3 MeV.

In the energy range of intermediate and thermal neutrons, resonance detectors are used which show distinct resonances at 580 keV (^{63}Cu) and 227 keV (^{65}Cu), respectively, 30 keV (^{23}Na), 4.8 eV (^{197}Au) and 1.4 eV (^{115}In), and are calibrated for a 1/E spectrum.

It is only in rare cases, when the reactor core is completely moderated, that the critical assemblies used for experiments are characterized by a leakage spectrum with a 1/E dependence [3]. Figure 1 shows calculated leakage spectra behind different shieldings. The calculation of these leakage spectra are based on the multi-diffusion theory and on a fission spectrum from an uranium rod of 4 cm diameter [4]. A 1/E spectrum is not obtained in this case either.

It is not known to what extent a neutron fluence measurement with resonance detectors will be in error in the absence of a 1/E spectrum, the more so, since the activation at the resonances takes place primarily in the eV range.

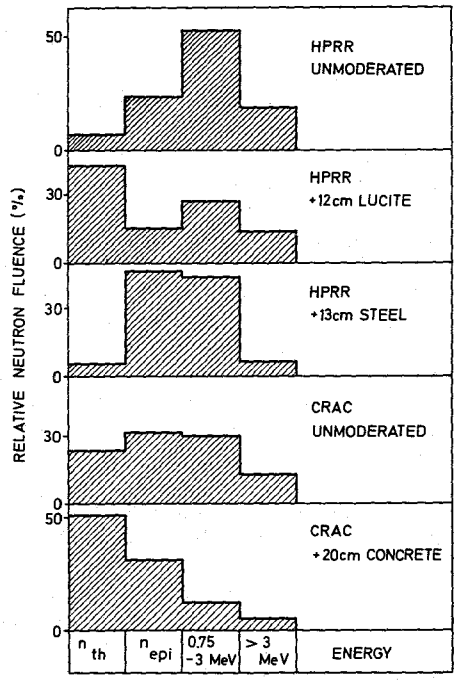


FIG.2. Neutron energy fluence histogram for neutrons from different critical facilities measured in air with moderators interposed.

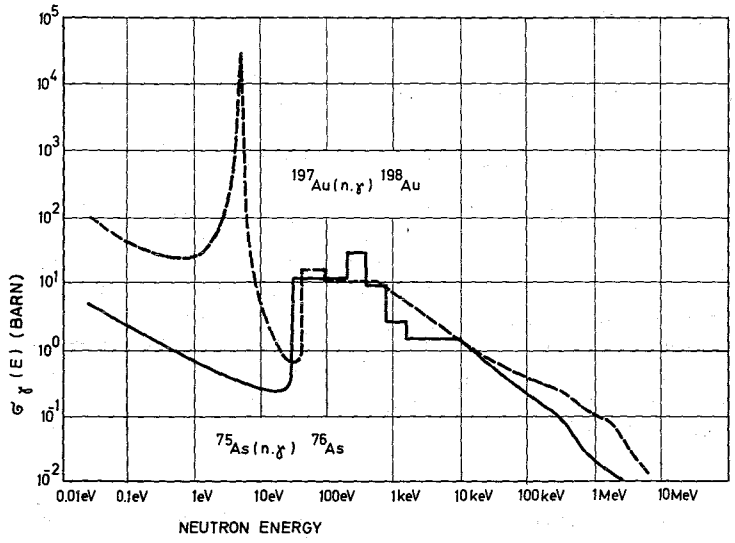


FIG.3. Neutron capture cross-section of arsenic and gold calculated in the resonance region from data of Ref.[9].

Since only intermediate neutrons above 1 keV contribute significantly to the total neutron dose, it is appropriate to use detectors providing an almost energy-independent indication over the range of intermediate neutrons and which, above all, are free of resonances below 100 keV. In search of an activation detector without distinct resonances we discovered arsenic to be a suitable detector [5].

Another aim in our work was to reduce the number of activation detectors in the conventional combinations and to improve the accuracy of measurement by detecting the activation fractions of thermal, intermediate and fast neutrons in a single detector and counting them in one single analyser. Transparent activation detectors, in particular one of arsenic phosphate glass, proved to be particularly useful.

A further task was to determine the direction of radiation incidence in a criticality accident. We used plastic belts fabricated of Makrofol E from which platelets can be punched out after irradiation. The microscopic counting of nuclear tracks produced directly in Makrofol E by recoil nuclei and (n, α) reactions allows a satisfactory determination of the dose distribution of fast neutrons around the person wearing this dosimeter belt.

This paper describes the results obtained with these detectors by intercomparison studies performed at different critical assemblies, namely:

A. The Health Physics Research Reactor (HPRR) at Oak Ridge, USA[1]: An unshielded unmoderated reactor loaded with enriched metallic uranium [6] (investigations conducted with the reactor unshielded, with a 13 cm Lucite shield and with a 13 cm steel shield);
B. The CRAC Irradiation Facility at Valduc, France[2]: A uranyl nitrate solution in a steel tank 30 cm in diameter [7] (investigations without shielding and with a 20 cm concrete shield).

The neutron fluence histograms which are typical for these critical assemblies are shown in Fig. 2.

2. ARSENIC AS AN ACTIVATION DETECTOR

2.1. Comparison of gold and arsenic activation

Arsenic has no resonance peaks within the energy range 1 MeV to 10 ke (cf. Fig. 3). Though in the energy range between 3 keV and 47 eV the differential cross-section for the neutron capture reaction $^{75}As\,(n, \gamma)^{76}As$ is characterized by a number of closely spaced resonances, the effective cross-section changes but insignificantly with neutron energy [8]. Below 40 eV the cross-section shows a $1/V$ dependence. The cross-section used for the range of resonances given was calculated using the Breit-Wigner formula and referring to the energy levels and the resonance integrals of arsenic given in Ref. [9]. When comparing the capture cross-section of

[1] Seventh Nuclear Accident Dosimetry Intercomparison Study, 20-31 July 1970, ONRL DOSAR, Oak Ridge, USA.

[2] IAEA Intercomparison Experiment and Research Co-ordination on Nuclear Accident Dosimetry, 15-26 June 1970, CEN Valduc, France under IAEA Research Agreement.

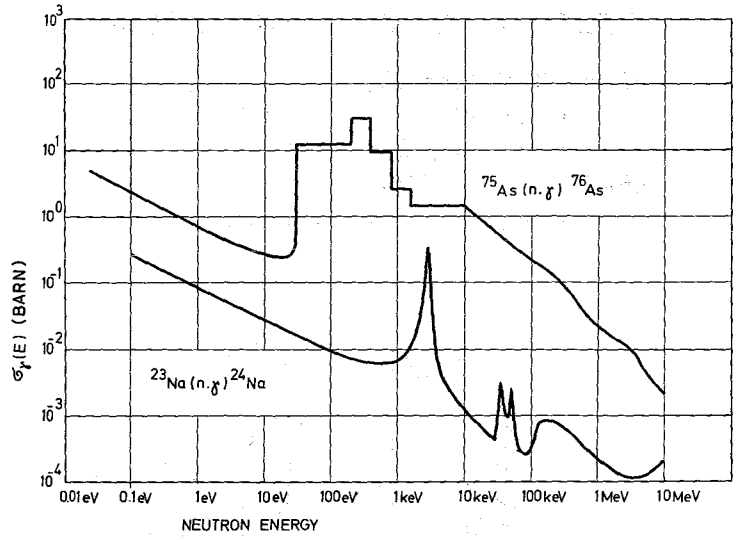


FIG.4. Neutron capture cross-section of arsenic and sodium from data of Ref.[9].

arsenic to that of the reaction $^{197}Au(n,\gamma)^{198}Au$, that of arsenic shows approximately the same sensitivity in the energy range above 40 eV but no resonances below 40 keV.

Further advantages are gained by using arsenic as a threshold detector for intermediate neutrons. For instance, a threshold energy of 40 eV is obtained when arsenic is covered by a suitable combination of cadmium, rhodium and gold foils.

There is another reason which points toward using the arsenic capture reaction for neutron measurement. The cross-section shows a curve similar to that of sodium without possessing the distinct resonances (see Fig. 4). Therefore a comparison is possible between arsenic activation in the personnel dosimeter and sodium activation in the body of the person exposed for an energy range which is of particular interest in dosimetry.

However, contrary to sodium, arsenic is characterized by: (a) the absence of resonances in the range 10 keV to 1 MeV; (b) a much greater cross-section (by the factor of about 1000); (c) a longer half-life of the activation product (^{76}As: $T_{\frac{1}{2}} = 26.4$ h); (d) a simple and sensitive measurement of β-activity in the transparent detector ($\beta_{max} = 3$ MeV).

Our preliminary investigations aimed to show the extent to which arsenic was applicable in accident dosimetry, as a substitute for other resonance detectors, such as sodium, gold or indium, for measurements of intermediate and thermal neutrons.

We used for our first experiments an As_2S_3 glass, 10 mm in diameter and 1 mm thick[3]. Irradiation took place with the arsenic glass bare, and simultaneously cadmium-covered gold foils were irradiated also; the values of neutron fluence were calculated from the neutron-induced β-activity.

---

[3] Manufacturer: Messrs Jenaer Glaswerke, Schott und Gen., Mainz, Federal Republic of Germany.

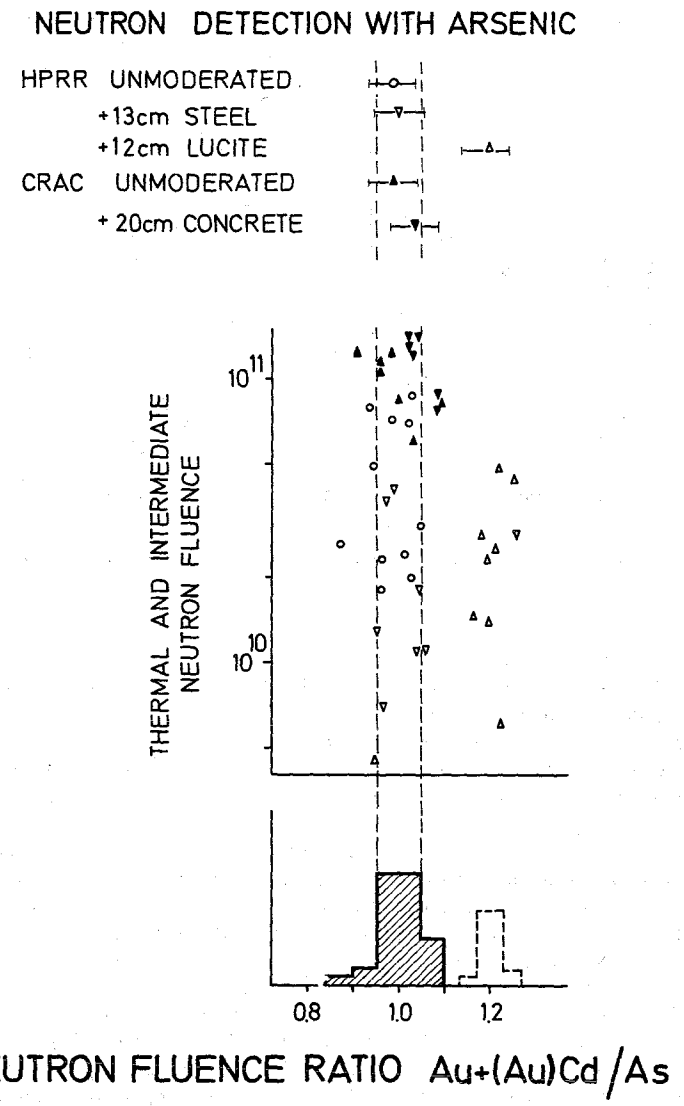


FIG.5. Comparison of neutron fluence (thermal and intermediate) measured with gold and arsenic at different critical facilities.

Figure 5 shows the ratio of thermal and intermediate neutron fluence measured with the gold and arsenic that was obtained for the different neutron leakage spectra, in particular in the case of phantom irradiation. The upper part of the figure shows the average values for one irradiation run, while the frequency distribution of the deviations of the measured values is represented in the lower part. On the whole, the agreement between gold and arsenic fluence values was found to lie within ±5%. Only in the case of phantom irradiation at HPRR shielded by 12 cm of Lucite was a greater

TABLE I. CHARACTERISTICS OF TRANSPARENT ACTIVATION DETECTORS

Detector	Neutron reactions	E_n (MeV)	Half-life $T_{\frac{1}{2}}$	β_{max} (MeV)	Count rate[a] per 10^{10} n/cm^2 (cpm)
Phosphate glass	$^{31}P(n,p)^{31}Si$	2.5	2.6 h	1.5	1840
8 mm × 8 mm × 4.7 mm	$^{31}P(n,\gamma)^{32}P$	0.1	14 d	1.7	11.5
As_2S_3 glass	$^{75}As(n,\gamma)^{76}As$	0.1	26.4 h	3	9600
10 mm diam. × 1 mm	$^{32}S(n,p)^{32}P$	2.9	14 d		
As phosphate glass	$^{31}P(n,p)^{31}Si$	2.5	2.6 h	1.5	1840
	$^{75}As(n,\gamma)^{76}As$	0.1	26.4 h	3	288
8 mm × 8 mm × 4.7 mm	$^{31}P(n,\gamma)^{32}P$	0.1	14 d	1.7	11.5

[a] Of slow or fast neutrons for burst time and Cerenkov measurement in a liquid scintillation counter.

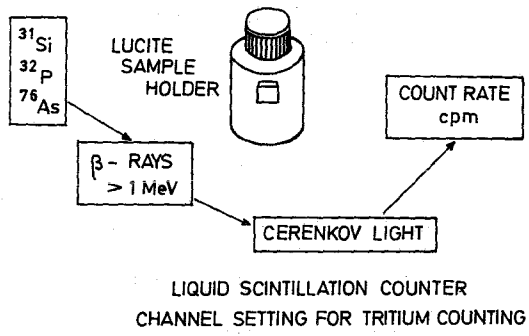


FIG. 6. Measuring technique of β-counting in transparent activation detectors.

deviation (+20%) observed. Calculations of the probability of arsenic capture and further studies with other neutron spectra would reveal to what extent this deviation is caused by the different cross-sections of gold and arsenic in the electron-volt energy range.

It appears from these results that arsenic can be regarded at least as an equal to gold as an activation detector for thermal and intermediate neutrons. Consequently, arsenic could replace conventional resonance detectors. The minor deviations of measured values from each other are negligible in dose measurement since, in practice, only neutrons above 1 keV contribute to the total neutron dose.

2.2. Properties of transparent arsenic phosphate glasses

In order to reduce the technical complexity we have used transparent activation glasses with success for some years [10, 11]. Characteristics are shown in Table I. The β-activity of these detectors can be determined

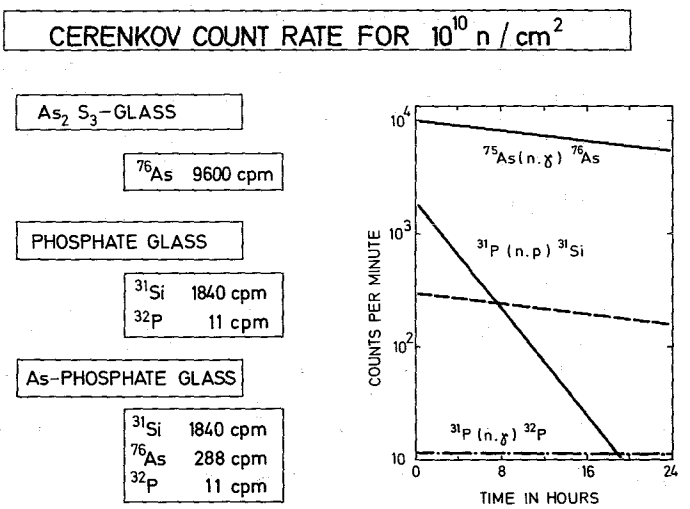


FIG.7. Count rate of ^{31}Si, ^{32}P and ^{76}As in arsenic and phosphate glasses measured in a liquid scintillation counter after irradiation with 10^{10}n/cm^2 (slow or fast neutrons).

easily by measuring the Cerenkov radiation induced by β-particles in the transparent glass (see Fig.6). The measurements are carried out in a liquid scintillation counter with the transparent glass in a sample holder of Lucite, and with the usual channel setting for tritium counting.

The advantages of this method include high counting efficiency, a good discrimination against disturbing lower-energy β-rays, a background count rate of about 10 cpm, simple transfer of calibration to other measuring equipment, and a separate measurement of the neutron fluence of fast and slow neutrons in the same glass and counter.

Figure 7 shows the Cerenkov counting rate of ^{31}Si, ^{32}P and ^{76}As obtained in a liquid scintillation counter after irradiation with 10^{10} n/cm^2. From this it can be seen that an As_2S_3 glass shows a relatively high sensitivity to thermal and intermediate neutrons.

The γ-dosimeter used for routine personnel monitoring at the Karlsruhe Nuclear Research Centre consists of a silver-activated phosphate glass[4] of 8 mm × 8 mm × 4.7 mm size contained in a boron-loaded plastic sphere, having a perforated tin filter of 2 mm thickness (energy dependence ± 8% over the energy range 45 keV to 1.2 MeV [12]). This phosphate glass is also used as a neutron activation detector for the detection of:

(a) fast neutrons above 2.5 MeV via the reaction ^{31}P (n, p)^{31}Si;
(b) thermal neutrons via the reaction ^{31}P (n, γ) ^{32}P.

To separate the two activation fractions ^{31}Si ($T_{\frac{1}{2}}$ = 2.6 h) and ^{32}P ($T_{\frac{1}{2}}$ = 14 d) from each other, a measurement is made by 15 hours (for ^{31}Si) and by 24 h (^{31}P) after irradiation.

[4] Yokota glass, FD-1.

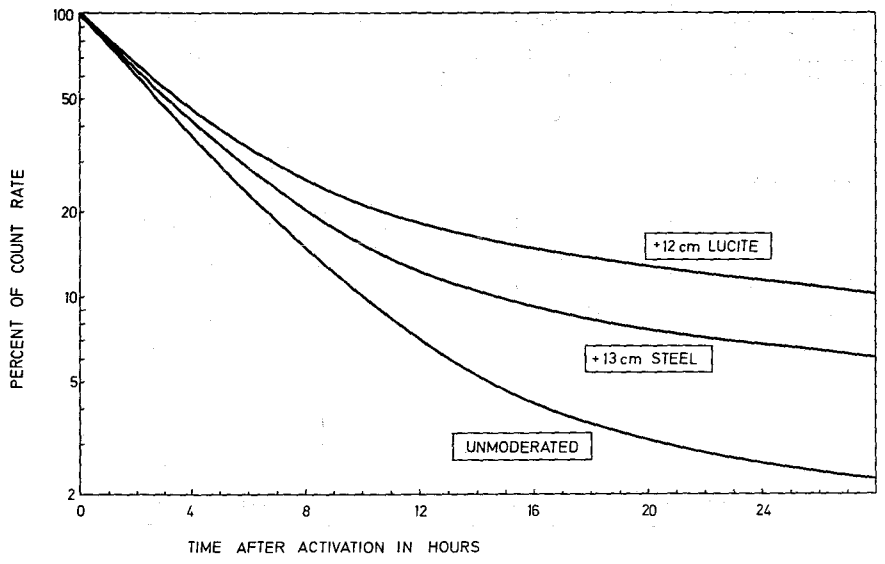


FIG. 8. The relative count rate of the arsenic phosphate glass as a function of the time after free-air irradiation measured for different neutron leakage spectra on the HPRR in Oak Ridge.

To improve the sensitivity of the phosphate glass to slow neutrons we developed, in cooperation with Jenaer Glaswerke, Schott and Gen., a newer arsenic phosphate glass, for a size of 8 mm × 8 mm × 4.7 mm, having a nominal arsenic weight fraction of 0.2%. In this case also, the different half-lives of ^{31}Si (2.6 h) and ^{76}As (26.4 h) allow separate measurements of the two activation fractions to be made. The relative slope of the counting rate for arsenic phosphate glass is shown in Fig. 8 after free-air irradiations at HPRR.

The results obtained from intercomparison irradiations showed that an arsenic phosphate glass may well be used to identify the shape of the neutron spectrum and that, under certain conditions, a direct measurement of the total neutron fluence or the total neutron dose will be possible.

2.3. Count rate ratio $^{31}Si/^{76}As$

If after irradiation two measurements are carried out, for example, after 6 and 24 hours, respectively, the count rate fractions of ^{31}Si and ^{76}As can be determined at time $t = 0$ (burst time). The count rate ratios $^{31}Si/^{76}As$ found for free-air and phantom irradiations at HPRR are listed in Table II and Fig. 9. For the irradiations carried out in the CRAC Facility, Valduc, the count rate ratio was calculated from the measured values for the As_2S_3 glass and for the phosphate glass.

The dosimeters irradiated in free air allow sufficient characterization of the neutron spectrum. It should be noted that a ratio of 6.8 of the measured values is obtained for the leakage spectrum behind the steel shielding, as compared to 3.7 and 1.2, respectively, for a hydrogen shielding. Thus, the counting rate ratio $^{31}Si/^{76}As$ can be used to correct the neutron dose in the energy range 0.7 - 2.5 MeV.

TABLE II. THE COUNT RATE RATIO SILICON-31/ARSENIC-76 MEASURED WITH THE ARSENIC PHOSPHATE GLASS FOR DIFFERENT NEUTRON LEAKAGE SPECTRA

Neutron spectrum	Count rate ratio = Si-31 / As-76			
	Free air dosimeter	Personnel dosimeter	Belt	
			Front	Back
HPRR				
Free air	21	2.5	2.4	1.8
+ 13 cm steel	6.8	2.6	2.0	1.7
+ 12 cm Lucite	3.7	2.3	2.7	1.7
CRAC				
Free air	4.1	2.3	2.2	0.5
+ 20 cm concrete	1.2	0.9	0.9	0.5

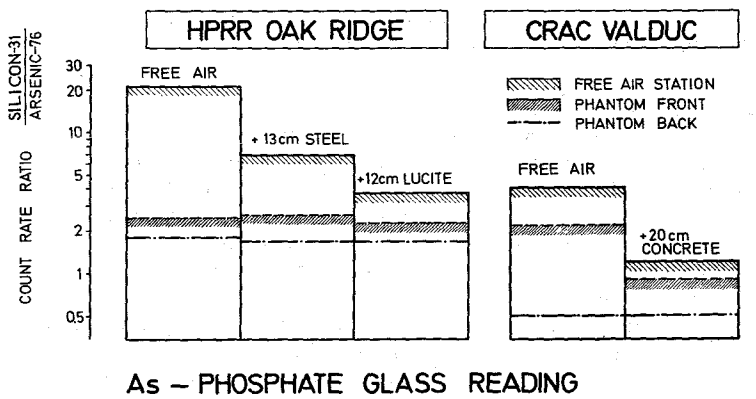


FIG. 9. The count rate ratio silicon-31/arsenic-76 measured with the arsenic phosphate glass on the free-air station and on the phantom for different neutron leakage spectra corrected for burst time.

Due to neutron moderation in the phantom, lower values are obtained for the count rate ratio which are almost independent of the spectrum, namely 2 to 2.7 and 0.9, respectively, for frontal irradiation of the dosimeters on the phantom and 1.7 and 0.5, respectively, for irradiation from behind.

2.4. Direct measurement of the total neutron fluence and neutron dose

With the arsenic content fixed in an appropriate way, use can be made of the more rapid slope of the ^{31}Si count rate portion as compared to the ^{76}As portion to indicate the total neutron fluence or the total neutron dose by a single count of the glass at a defined moment after activation.

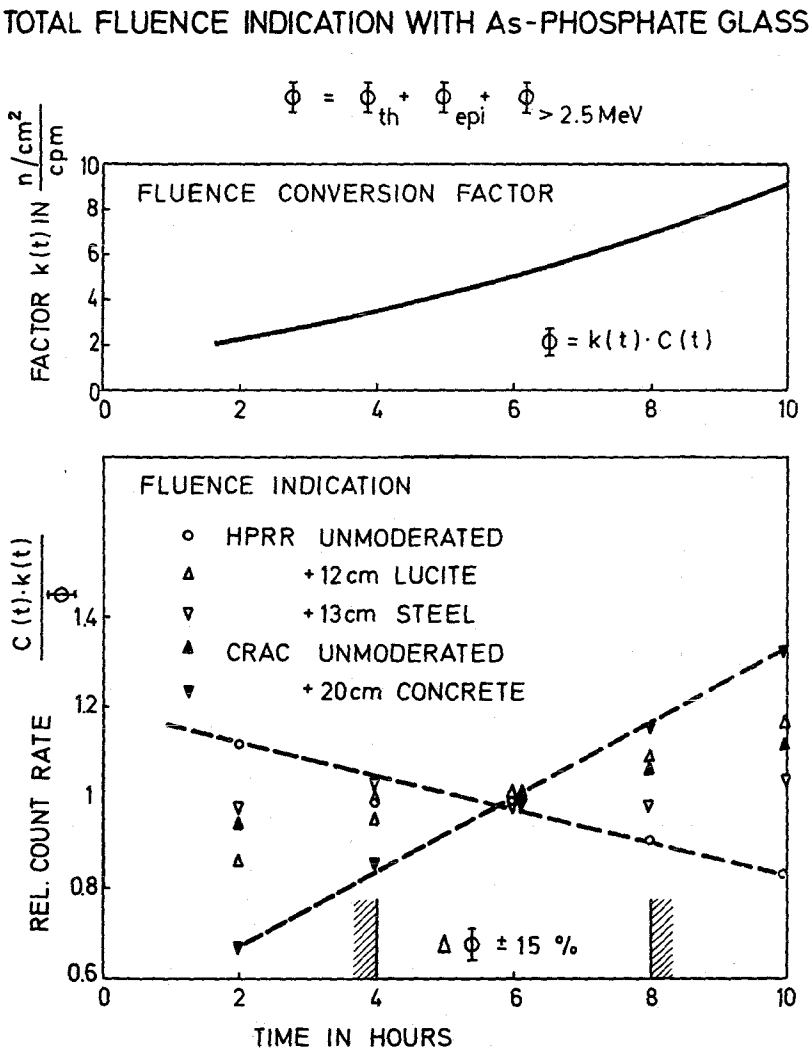


FIG.10. The fluence conversion factor and the direct indication of the total neutron fluence measured with the arsenic phosphate glass in the first hours after the burst for different neutron leakage spectra.

As an example, the arsenic phosphate glass used in the experiments provides direct indication of the neutron fluence ($\Phi = \Phi_{th} + \Phi_{epi} + \Phi_{2.5\,MeV}$) 6 hours after activation of the glass. However, using a fluence conversion factor k(t) which is dependent on the moment of activation, the measurement may also be performed within an interval of 4 to 8 hours after activation. These conditions are shown in Fig. 10. Independent of the neutron spectrum obtained, the count rate measured at time t= 6 hours is directly proportional to the total neutron fluence. In the time interval 4 to 8 hours after activation a maximum inaccuracy of ± 15% would result, depending on the

TABLE III. PRELIMINARY RESULTS OF SEVENTH INTERCOMPARISON OF NUCLEAR ACCIDENT DOSIMETRY SYSTEMS AT HPRR, OAK RIDGE

Location	Participants	First collision dose (rad)					
		Burst I		Burst II		Burst III	
		n	γ	n	γ	n	γ
Free-air station	ORNL (DOSAR)						
	TDU-dosimeter	302	44.6	37.7	35	109	11.2
	Proportional counter	318	--	37	--	118	--
	Range of all participants	287-351	34-113	37-79	22-36	73-154	5.4-14
	KARLSRUHE GFK						
	preliminary results	302	43.5	54	36	132	8
	corrected results	300	46	38	33	115	10.5
Irradiation with phantom	ORNL (DOSAR)						
	(from ^{24}Na)	315	--	43	--	111	--
	Range of all participants	257-433	59-113	36-67	39-57	73-192	27-34
	KARLSRUHE GFK						
	preliminary results	355	93	65	57	143	33
	corrected results	348	86	46	53	128	27
	belt front	349	78	42	56	136	29
	belt back	40	50	15	24	23	20

BURST I: HPRR unmoderated
BURST II: + 12 cm Lucite
BURST III: + 13 cm steel

neutron spectrum. Accordingly, a total neutron dose ($D = D_{th} + D_{epi} + D_{> 2.5\ MeV}$) can also be indicated in a direct fashion. This is possible, for example, with an arsenic phosphate glass whose arsenic content is only about 1/4. The sensitivity obtained for the first-collision dose 6 hours after activation is about 13 cpm/rad both for slow neutrons (^{76}As) and for fast neutrons (^{31}Si). Consequently, two counts of the arsenic phosphate glass allow the separate determination of both dose fractions (Si, As), while direct indication of the total dose is provided when counting only once at a defined moment after activation.

When, for calibration of the phosphor threshold detector, an unmoderated fission spectrum or the HPRR spectrum is used, an arsenic phosphate glass will indicate the total dose over the entire energy range.

However, a moderation of the spectrum must be taken into account by an energy conversion factor. This factor allows for the relative change of the fluence fraction within the energy range 0.75 to 2.5 MeV. The assumptions regarding the spectrum shape are based on the count rate ratio $^{31}Si/^{76}As$ measured with the free-air dosimeter.

After a criticality accident the phosphate glass, both γ-dosimeter and neutron activation dosimeter, allows evaluation of the γ-dose and, to a first approximation, a sufficiently accurate determination of the neutron dose. Experimental results obtained with the phosphate glass dosimeter and the arsenic glass during the 7th Nuclear Accident Dosimetry Intercomparison Study at Oak Ridge are shown in Table III. The table gives a comparison of our results with preliminary results from all participants in the Study, and the results obtained with the TDU-dosimeter, a phosphate glass dosimeter (measurement of γ-dose), and a proportional counter of the ORNL Dosar Facility in Oak Ridge. Using the ratio $^{31}Si/^{76}As$ obtained from the arsenic phosphate glass, the phosphate glass dose for fast neutrons was corrected (by a factor of 2.5) with respect to the irradiation shielded by 13 cm of steel.

3. MAKROFOL-E AS A SOLID-STATE NUCLEAR TRACK DETECTOR

3.1. Properties of the detector

For detection of fast neutrons above a threshold of 0.75 MeV, 1.3 MeV and 1.5 MeV a combination of Makrofol E[5] with neptunium, thorium and uranium, respectively, is used. However, fast neutrons can also be detected directly by recoils and (n, α) reactions in carbon and oxygen in a Makrofol E plastic foil. Makrofol E must be regarded as the simplest solid-state nuclear track detector. Compared with Triafol, the material characteristics of Makrofol E are preferable (greater hardness, no environmental influences, homogeneous material with possibilities of getting reproducible measurements).

After irradiation the 300 μm Makrofol E foils are etched at 60°C in 35% KOH solution. As compared with the procedure used for fission fragment tracks, a longer etching of about 4 hours is required in this case (Fig. 11).

---

[5] Polycarbonate foil from Bayer AG., Leverkusen, Federal Republic of Germany.

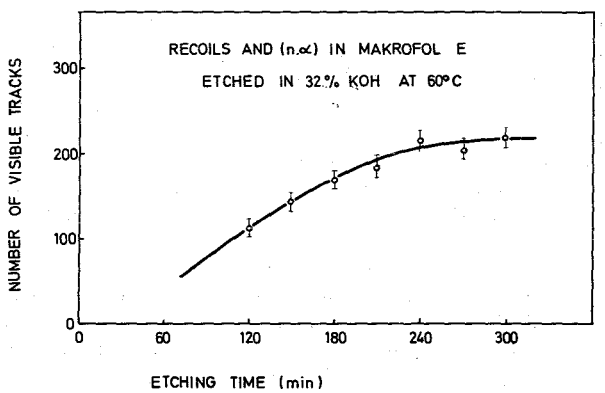


FIG. 11. The relative number of visible etch pits from recoils and α-particles in Makrofol E as a function of the etching time in 32% KOH at 60°C.

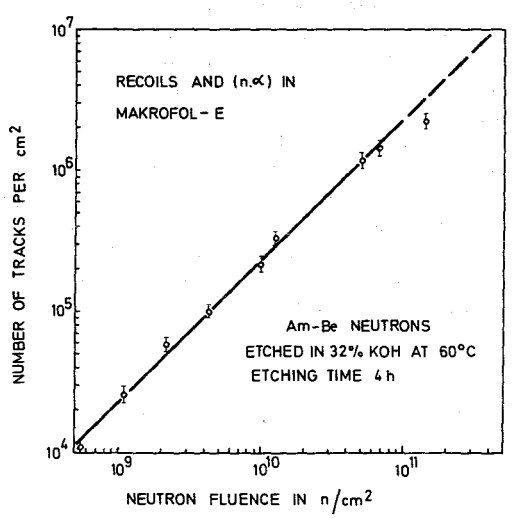


FIG. 12. The number of visible recoil and α-particle tracks in Makrofol E as a function of the neutron fluence (AmBe neutrons).

Direct detection of neutrons in Makrofol E is a very sensitive method; contrary to the case for fission fragments, the etch pits of recoils and α-particles are different in size and therefore less well suited for nuclear track counting; this may result in individual calibration values of some (1 to 2) $\times 10^{-5}$ tracks per neutron for AmBe neutrons (see also [13]). Figure 12 shows the recoil and α-particle tracks per square centimetre counted under the microscope as a function of the neutron fluence. Nuclear track counting is possible over some 4 decades. This corresponds to a dose range above 1 rad up to several 100 rad.

The detector combinations containing electrolytically deposited neptunium, thorium or uranium furnish 10^{-6} to 10^{-8} tracks/neutron depend-

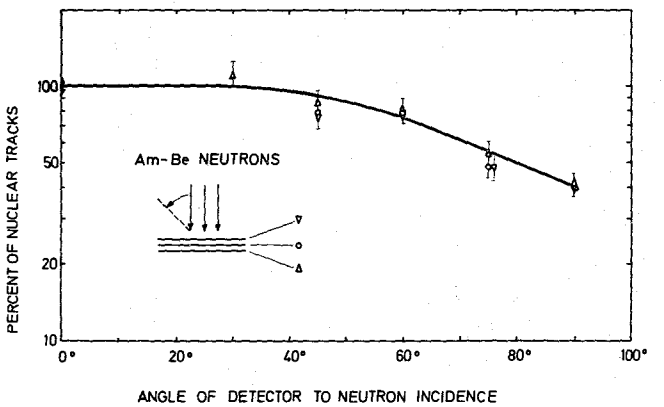


FIG. 13. The relative number of recoil and α-particle tracks in Makrofol E as a function of the detector angle to the direction of radiation incidence.

ing on the thickness of the layer (for thick metal foils with saturation layer 1.16×10^{-5} fission fragment tracks/(neutron · barn) [14]).

Due to the completely different image of the etching pit an individual microscopic counting of recoil and α-particle tracks in Makrofol E does not provide the same reproducibility as the counting of fission-fragment tracks. The reproducibility was $\pm$ 10%, found for relative measurements.

Makrofol E is particularly resistant to the influences of temperature and air humidity. Unlike cellulose-nitrate [15], foils of Makrofol E show practically no fading up to a temperature of 100°C and a relative humidity of 100%. Effects due to environmental influences during irradiation are not observed.

The detection of recoil and α-particle tracks in Makrofol E is dependent on the direction of the incidence of the radiation (Fig. 13). When irradiation is parallel to the film surface only 40% of the tracks are detected. This condition must be taken into account for accurate fast neutron dosimetry.

3.2. Nuclear track counting

The difficult conditions obtaining in nuclear track counting of recoils and α-particles will be explained for different etch pits. Figure 14 shows etch pits of fission fragments in mica and in Makrofol E. Here, etch pits clearly contrast with the background of the picture. Our work has shown that by using the new Interference-Contrast Technique of Nomarski[6] an additional contrast can be achieved by producing different interference colours (yellow, red, blue) in the etch pits to those in the background of the image[7] [16, 17].

---

[6] Manufacturer: Messrs Carl Zeiss, Oberkochen, Federal Republic of Germany.

[7] The effect of these photomicrographs is considerably enhanced by the colouring; however, colour reproductions could not be included in these Proceedings. Readers interested can find examples in Refs [16, 17].

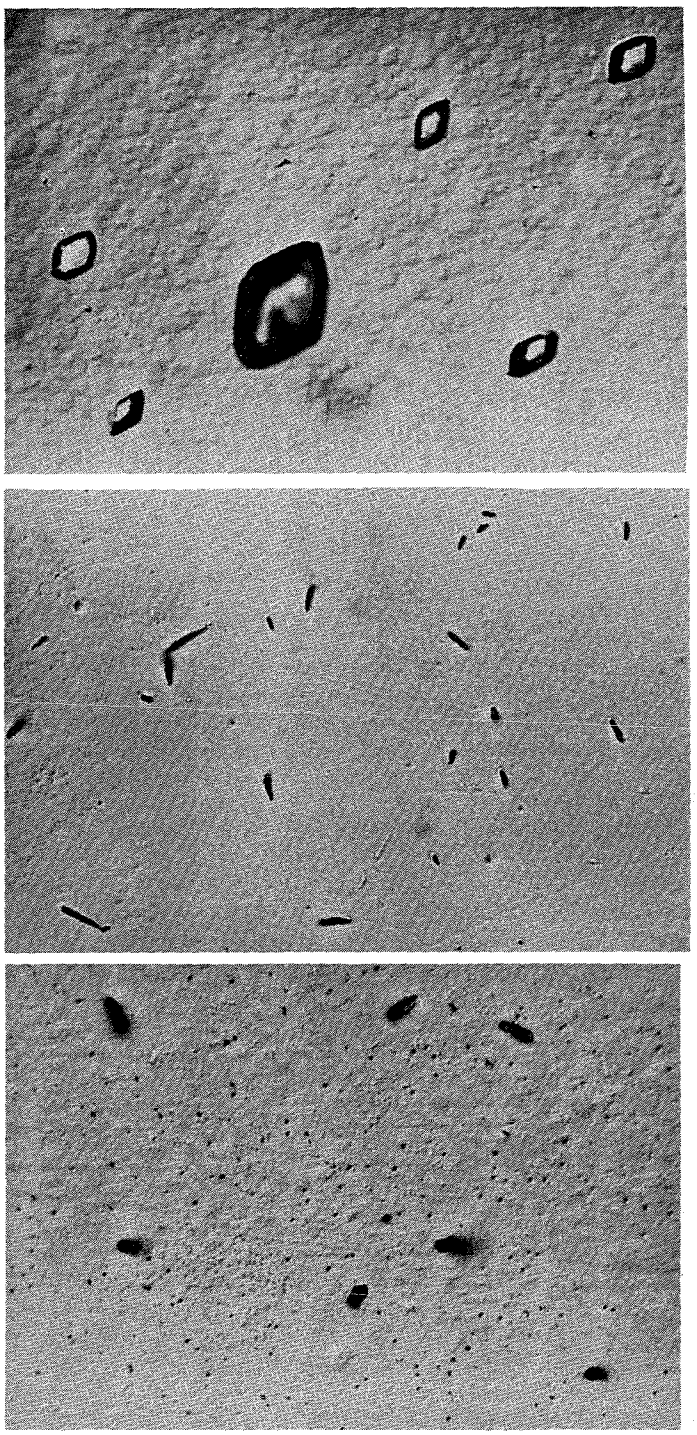


FIG. 14. Photomicrographs (~×550) of fission fragment tracks after neutron irradiatio in contact with uraniur (see footnote 7): (a) In mica after 1 h etching in HF and H_2SO_4 at 60°C (pre-etching of natural fission fragment tracks before neutron irradiation); (b) In Makrofc E after etching in 30% KOH at 60°C, etching time 30 min; (c) In Makrofol E after an etching time of 2 hou (small etch pits from α-particles).

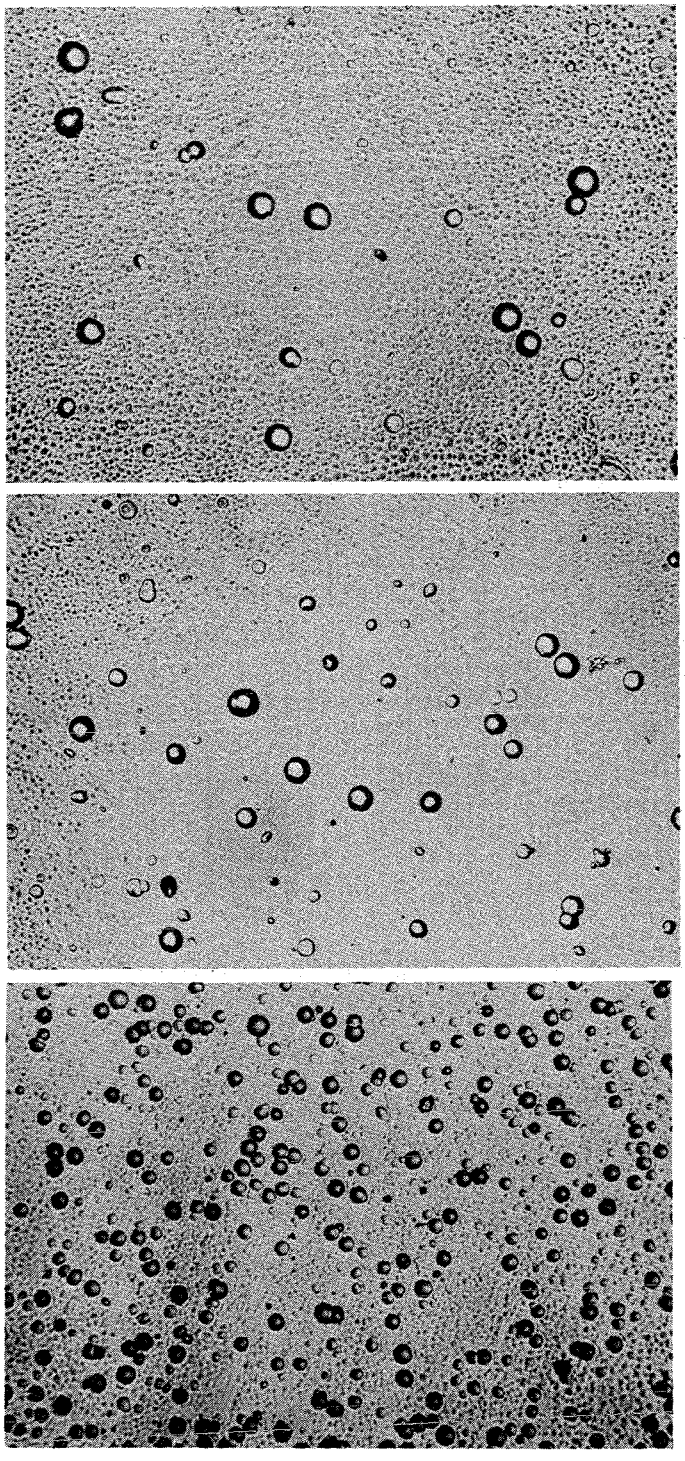


FIG. 15. Photomicrographs (~× 550) of recoil and α-particle tracks in Makrofol E exposed to neutrons without an additional radiator after 4 hours etching in 30% KOH at 60°C (Interference Contrast Microscopy) (see footnote 7): (a) etch pits (2 to 10 μm diam.) with background after exposure to 14 MeV neutrons; (b) etch pits after background discrimination by means of a colour contrast; (c) etch pits (2 to 7 μm diam.) after exposure to fission neutrons.

The detection of recoils and α-particles in Makrofol E requires a somewhat delicate counting technique. This is mainly due to the different sizes of pits but also, on account of the long etching time, to the background, which is porous, in particular at high track densities (Fig. 15).

In spite of the long etching time, tracks formed by recoils and α-particles have small diameters ($2-7\,\mu$m). The microscopic counting of several hundreds of tracks in each field of view places an excessive demand on the operator. It is therefore necessary to divide the field of view and to count the photographic picture if a sufficient counting reproducibility is to be obtained.

However, individual counting of nuclear tracks under the microscope is a very tedious procedure even with recourse to the most recent optical contrast techniques; besides, it is inadequate with both a low or a very high density of tracks.

Measurement of the optical density cannot be applied to the low track densities in personnel dosimetry, but at higher exposures it can be used satisfactorily, in particular with relative measurements [18].

A remarkable innovation, developed by Cross [19], was automatic counting of etch pits. The counting involves generation of electrical discharges across the pits in thin polycarbonate foils; the discharge is between an aluminized Mylar foil and a thick metal electrode at a sparking potential of ~ 600 V. We were successful in using this method to count fission fragments of neptunium, uranium and thorium in a 12 μm Makrofol E foil. The possibility of counting large areas (several square centimetres) results in a considerable sensitivity (lower limit of detection about 1 track/cm^2). Since the replicas of the etch pits are shown 20 times enlarged by aluminium evaporation onto the Mylar foil, a range of only four decades is covered here. Having cleaned the foil carefully (ultrasonic washing before etching; drying in a dessicator) and using non-corroding materials for the electrodes, e.g. stainless steel, we obtained counting accuracies of $\pm 10\%$.

The same method is applicable to the counting of α-particles in thin cellulose nitrate foils [20]. Our investigations have shown that with the commercially available foils of Makrofol E the counting of neutron-induced recoil and α-particle tracks is not practically feasible due to the lack of reproducibility and a relatively high background of tracks in the unirradiated foil.

Automatic counting of the microscopic image, possible with recent electronic counting systems (Quantimet, Integramat, Mikro-Videomat[8]), is a very promising technique; however, considerable difficulties arise when counting high track densities or large detector areas. It is true that the transmission of the microscopic image to a television screen offers additional possibilities of achieving good contrast, as well as a discrimination against small background tracks and a separate counting of tracks of different sizes. However, larger, i.e. particularly profound, etch pits might be overlooked in automatic counting, and the discrimination against smaller tracks depends on the individual contrast setting used. We think that a combination of the optical Interference-Contrast Technique and an

[8] Manufacturers: Quantimet – Messrs Metals Research, England.
Integramat – Messrs Leitz, Wetzlar, Federal Republic of Germany.
Micro-Videomat – Messrs Carl Zeiss, Oberkochen, Federal Republic of Germany.

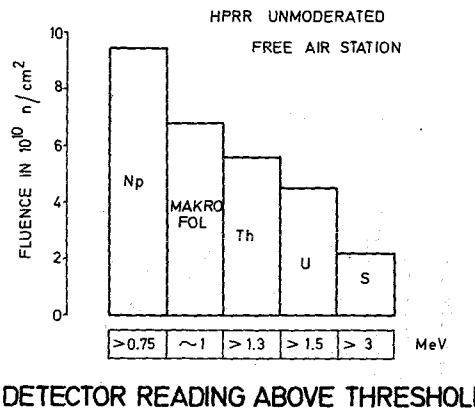


FIG.16. The fast neutron fluence at the unmoderated HPPR measured with Makrofol E and different threshold detectors. Neptunium, thorium and uranium exposed in contact with Makrofol E; all detectors were calibrated to AmBe neutrons.

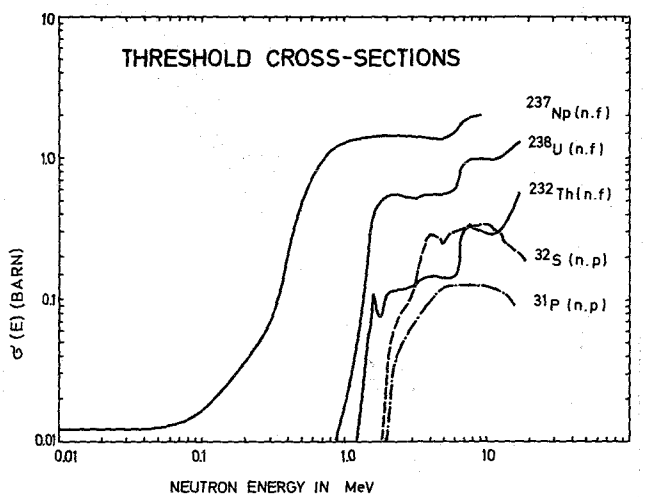
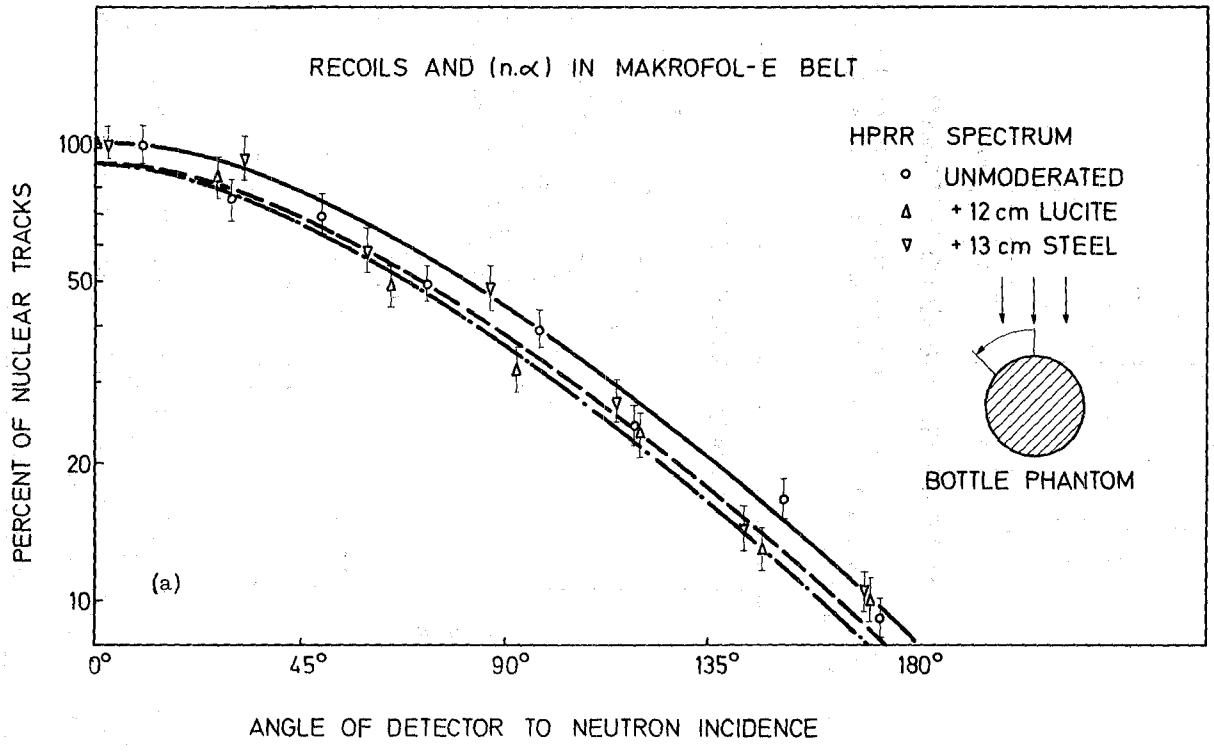


FIG.17. The cross-section of different threshold detectors for fast neutrons as a function of energy.

automatic counting with the Mikro-Videomat will provide further possibilities of improving the reproducibility of a neutron measurement with Makrofol E (in particular as regards absolute measurement) as compared with actual individual track counting.

3.3. Threshold detector characteristics

The knowledge of the threshold energy is important in the practical application of a Makrofol E detector. First data [13] were based on calibration exposures with neutrons of 14 MeV, AmBe and (d, D) neutrons,


RECOILS AND (n.α) IN MAKROFOL-E BELT
HPRR SPECTRUM
UNMODERATED
+12 cm LUCITE
+13 cm STEEL
BOTTLE PHANTOM
PERCENT OF NUCLEAR TRACKS
100
50
20
10
(a)
0°
45°
90°
135°
180°
ANGLE OF DETECTOR TO NEUTRON INCIDENCE

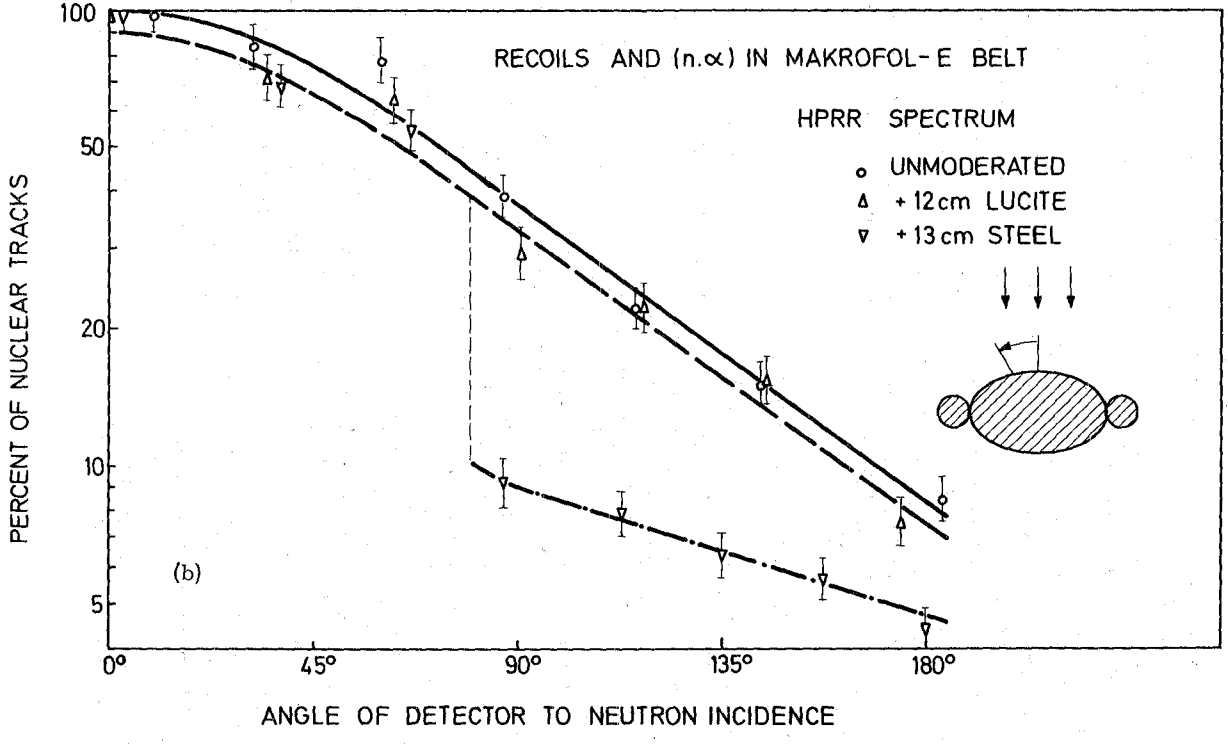


FIG.18. The relative number of recoil and α-particle tracks in Makrofol E as a function of the detector angle to the neutron incidence at the HPPR: (a) Makrofol E belt exposed on a bottle phantom; (b) Makrofol E belt exposed on a phantom of elliptical cross-section.

as well as HPRR fission neutrons. Since, in elastic scattering, neutrons transfer low energies to carbon and oxygen recoil nuclei, the smallest detectable neutron energy was estimated to be in the energy range 0.5 - 1.0 MeV. More accurate calibrations with monoenergetic neutrons in the energy range of 1 MeV and below are not yet available. Newer values were published for Triafol T [21]; here the energy dependence of the response indicates a reasonably good agreement to the first collision dose in the energy range 1 to 14 MeV, and also for monoenergetic 1.2 and 0.5 MeV neutrons.

We irradiated Makrofol E foils at HPRR together with sulphur and with detector combinations consisting of Makrofol E in contact with electrolytically deposited neptunium, thorium and uranium. The counting results of these detectors, which were calibrated to AmBe neutrons, furnished the data listed in Fig.16. A comparison with the threshold energies of 0.75 MeV, 1.3 MeV, 1.5 MeV and 2.9 MeV determined from the known cross-sections (Fig.17) shows that the threshold energy for the recoil and α-particle track counting of Makrofol E lies between 0.75 and 1.2 MeV, most probably around 1 MeV.

3.4. Phantom studies with Makrofol E

For testing the practical applications in accident dosimetry, irradiations of a phantom were carried out using a plastic belt of Makrofol E at HPRR, Oak Ridge. The belt is fabricated of a 5 cm wide Makrofol strip welded into a thinner Makrofol foil. After the irradiation discs of 1 cm diam. are punched out of the belt at uniform distances, etched and counted under a microscope. Figure 18 shows the relative dose indication of the Makrofol detector for the case of frontal irradiation of the phantom as a function of the position of the dosimeter with respect to the angle of radiation incidence. The drop of dose indication in the range from 0° to 90° is mainly due to the directional dependence of the detector foil (cf. also Fig.13). The influences due to the shape of the fission spectrum and the form of the phantom are smaller than $\pm$ 10% in the case considered. With the cylindrical phantom only was the indication of scattered neutrons lower for the HPRR spectrum moderated by 13 cm of steel, and this was probably due to the influence of the energy threshold below 1 MeV.

A belt of Makrofol E is a simple detector for determining the direction of the incident radiation after a criticality accident. Our irradiations of phantoms indicated that a remarkably strong effect is produced by the arms or other scatterers near the body, in particular for radiation incident at 90° (cf. Figs 19, 20). The use of two dosimeters instead of a dosimeter belt is obviously inadequate for determining the direction of the incident radiation with any reasonable accuracy, the more so, since a correction of up to a factor of 10 of the personnel dosimeter indication is necessary for the case of irradiation from behind.

4. SUMMARY

The results obtained by intercomparison measurements at HPRR in Oak Ridge and in the CRAC Facility at Valduc served as a basis for describing the properties of an arsenic phosphate glass dosimeter for measurements of neutron dose, and the results derived from phantom studies with

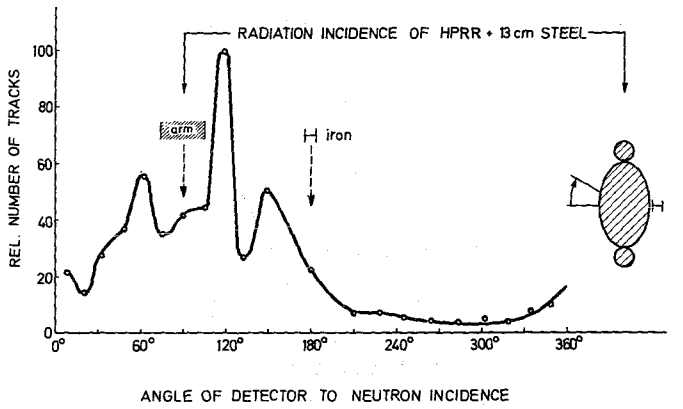


FIG.19. Study with phantom, wearing a belt of Makrofol E at the HPRR in Oak Ridge for a phantom irradiation at 90°. The relative number of recoil and α-particle tracks in Makrofol E is reduced by the influence of the phantom arm.

a Makrofol E belt to obtain a determination of the direction of the incident radiation.

When suitably calibrated, the arsenic phosphate glass allows the simultaneous detection of thermal and fast neutrons. Compared with the methods of measurement used at present, the following improvements have been recorded;

(a) Preferred fluence measurement of intermediate neutrons above 40 eV with arsenic replacing conventional resonance detectors;
(b) Separation of the two activity fractions, ^{76}As and ^{31}Si, by two measurements made within 24 hours of a criticality accident;
(c) Information on the type of moderation and shielding of the critical assembly furnished by the count rate ratio of $^{31}Si/^{76}As$ for the case of free-air irradiation;
(d) Direct indication of total neutron fluence or total neutron dose by direct measurement of the glass at a given moment after activation (e.g. 6 h); this depends on the arsenic content of the glass.
(e) A possible statement as to the orientation of the person wearing the dosimeter to the irradiation and, hence, obtaining an estimate of the real neutron dose by comparing the ^{76}As activity of the personnel dosimeter with the ^{24}Na activity in the blood.

The arsenic phosphate glass dosimeter can be reasonably and simply supplemented by using a solid-state nuclear track detector that consists of a single Makrofol E foil. The energy threshold for detection of neutrons via recoils and α-particles in Makrofol E is about 1 MeV, which means that, except for neptunium-237, the threshold is more advantageous than that of other, more expensive threshold detectors (^{115m}In, ^{232}Th, ^{238}U). Due to the useful threshold energy and the stability in storage, Makrofol E can usefully improve the information obtainable from the phosphate glass measurements.

In a criticality accident, a belt of Makrofol E could be the simplest, cheapest and lightest neutron detector for obtaining a determination of the orientation of the source with respect to the person exposed.

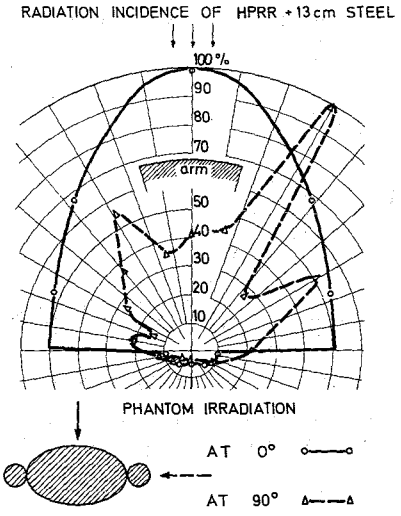


FIG. 20. Study with phantom, wearing a belt of Makrofol E at the HPRR in Oak Ridge for two different directions of radiation incidence.

Further intercomparison measurements and phantom studies will serve to evaluate the possibilities of criticality dosimetry based on a single dosimeter, in particular with regard to body orientation with respect to the source.

ACKNOWLEDGEMENTS

The author wishes to thank Dr. W. Jahn, Jenaer Glaswerke, Schott und Gen., Mainz, Federal Republic of Germany for the preparation of arsenic glasses, Dr. Comper and Mrs. I. Hofmann for their part in the research and development. Thanks are due also to Dr. J.A. Auxier and Dr. F.F. Haywood for the use of the Health Physics Research Reactor at Oak Ridge National Laboratory.

REFERENCES

[1] HURST, G.S. et al., Rev. Sci. Instrum. 27 (1956) 133.
[2] KERR, G.D., STRICKLER, T.D., Health Phys. 12 (1966) 1141.
[3] HANKINS, D.E., USAEC Rep. LA-3910 (1968).
[4] COMPER, W., unpublished work.
[5] PIESCH, E., Atompraxis, Direct Information 3/70 (1970).
[6] AUXIER, J.A., Health Phys. 11 (1965) 89.
[7] CANDES, P., LAMBERIEUX, J., "Dosimétrie pour les cas d'accidents nucléaires", Nuclear Accident Dosimetry Systems (Proc. Panel Vienna, 1969), IAEA, Vienna (1970) 25.
[8] COMPER, W., PIESCH, E., Nucl. Instr. Methods (1971).
[9] BROOKHAVEN NATIONAL LABORATORY, Neutron Cross-Sections, Second ed., USAEC Rep. BNL-325 (1966).

[10] PIESCH, E., "Neutron dose measurements by means of the Cerenkov effect of induced beta radiation in gamma and activation-dosimeters. (Proc. Symp. Vienna, 1966), IAEA, Vienna (1967) 471.
[11] MAUSHART, R., PIESCH, E., "Use of phosphate glasses for dosimetry of mixed radiation fields in neutron radiobiology", Solid State Chemical Radiation Dosimetry in Medicine and Biology (Proc. Symp. Vienna, 1966) IAEA, Vienna (1967) 157.
[12] PIESCH, E., Proc. 2nd Conf. Luminescence Dosimetry, Gatlinburg, Tenn. (1968) 830.
[13] BECKER, K., Proc. ENEA Symp. External Radiation, Stockholm (1967) 317.
[14] PRETRE, S., TOCHILIN, E., GOLDSTEIN, N., Proc. First IRPA Congress, Rome, Pergamon Press (1968) 491.
[15] BECKER, K., Health Phys. 16 (1969) 113.
[16] PIESCH, E., Proc. Int. Topical Conf. on Nuclear Track Registration, Clermont-Ferrand (1969).
[17] PIESCH, E., Zeiss Informationen 18. Jahrg. No. 76 (1970) 58.
[18] TUYN, J.W.N., Proc. Int. Topical Conf. on Nuclear Track Registration, Clermont-Ferrand (1969).
[19] CROSS, W.G., Proc. Int. Topical Conf. on Nuclear Track Registration, Clermont-Ferrand (1969).
[20] JOHNSON, D.R., BECKER, K., USAEC Rep. ORNL-TM-2826 (1970).
[21] BECKER, K., Proc. Int. Topical, Conf. on Nuclear Track Registration, Clermont-Ferrand (1969).

DISCUSSION

T.F. JOHNS: Could you please say a little more about the practical aspects of using the Makrofol belt? For example, what is the cost? Is it durable, that is, can one wear such a belt day by day for years without it deteriorating?

E. PIESCH: Makrofol is a commercial product produced by Bayer AG. Small quantities can be obtained free of charge. The dose indication seems to be very stable. But for the time being this system can only be used for relative and not for absolute measurements. Up to the present we have no experience of wearing such a belt, but life-testing, probably under the wearer's clothes, are foreseen. The belt is insensitive to temperature and humidity variations, and can be washed in boiling water.

C.O. WIDELL: I would like to mention another method for fast neutron dosimetry in cases of accident. The fast-reading dosimeter uses a pin junction silicon diode in which the rise in the forward voltage is measured.

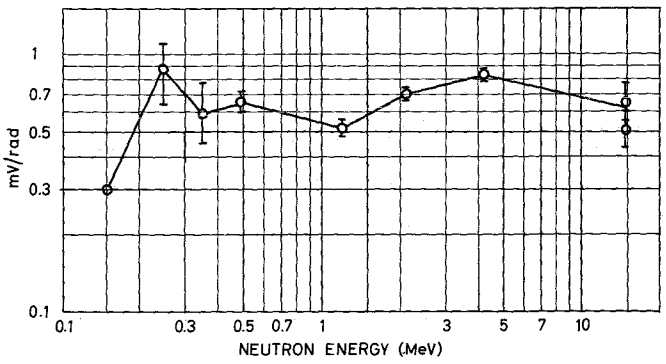


FIG.A. Differential sensitivity to neutrons of different energies. Each point represents the mean value for 4 diodes (Differential sensitivity at 25 mA and 25°C).

By means of this dosimeter it is possible to measure fast neutrons above 0.15 MeV down to 1 rad. Under our reading conditions the sensitivity is 0.7 (±20%) mV per rad above 0.15 MeV (see Fig. A). Furthermore, this detector can be read a long time after the irradiation without risk of appreciable fading.

G. BURGER: Further to the last statement, we have found that when using pure Makrofol at radiation levels where spark counting is of advantage, the relative standard deviations for single measurements amount to as much as 40%. The trouble is that by using only the carbon recoils one does not get a plateau for the sparks versus etching time, but rather an exponential increase. Until we have better foils, this disadvantage will not be overcome.

Now a question: what significance do you attach to your Fig. 19?

E. PIESCH: The results I have presented are for relative measurements. All Makrofol foils of one run were etched at the same time and counted for the same time under the microscope. Under these conditions we have obtained a standard deviation of ±10% for the direct detection of neutrons via recoils and α-particles. The results are given in Fig. 18, and those of Fig. 19 were obtained with the same significance.