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E. Piesch

GESELLSCHAFT FÜR KERNFORSCHUNG M.B.H.

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NEUTRON DOSE MEASUREMENTS BY MEANS OF THE CERENKOV EFFECT OF INDUCED BETA RADIATION IN GAMMA AND ACTIVATION DOSIMETERS

E. PIESCH
KARLSRUHE NUCLEAR RESEARCH CENTRE,
FEDERAL REPUBLIC OF GERMANY

Abstract

NEUTRON DOSE MEASUREMENTS BY MEANS OF THE CERENKOV EFFECT OF INDUCED BETA RADIATION IN GAMMA AND ACTIVATION DETECTORS. Activation techniques are often used in neutron monitoring, namely in criticality accidents, in the measurement of high-energy neutrons and of high-level neutron flux densities. The β-activity induced in activation detectors (reactions $^{32}S(n,p)^{32}P$, and $^{12}C(n,2n)^{11}C$) will be detected directly, after concentration or chemical separation of the sample by GM counters, proportional counters or liquid and plastic scintillators.

The possibility to measure the Cerenkov radiation of the β-radionuclide directly in the sample has the following advantages:

To determine a β-activity directly in the liquid phase or in a solid-state detector
To measure without absorption in the sample
To use a counting method with optimum detection efficiency.

The application of the Cerenkov radiation measurement will be described for activation detectors using the $^{32}S(n,p)^{32}P$ and $^{31}P(n,p)^{31}Si$ reactions for measuring fast neutrons, as well as for gamma detectors, especially silver-activated metaphosphate glass dosimeters (radiophotoluminescence dosimeters) suitable for measuring fast neutrons by the reactions $^{31}P(n,p)^{31}S$, and thermal neutrons by the ^{32}P-activity.

The advantages of these measurement techniques and the possibility of their practical application in personnel and area monitoring will be discussed.

1. INTRODUCTION

For the detection of fast neutrons by the activation of threshold detectors, conventional measuring methods are applied to measure the induced beta activity. The beta particles emitted by the sample are either measured directly or the activity of the residue is determined after prior chemical preparation of the sample to separate the radionuclides formed from the inactive substance [1-4]. The disadvantages of this method, which are particularly evident in routine evaluation of numerous samples or under the pressure of time in an accident, are either the relatively low measuring sensitivity due to the unfavourable measuring geometry and the high self-absorption in the carrier substance of the activation sample, or the tedious and expensive preparation of the sample with its inherent possibilities of error and mix-up.

The direct measurement of beta activity by means of the Cerenkov radiation generated in transparent activation detectors offers some important advantages, namely:

(a) The measuring geometry, which may be an optimum under favourable conditions, results in a better efficiency and, hence, shorter measuring time of the activation sample without previous preparation.

(b) The background counting rate of the measuring device, which may be lower under favourable conditions, enables the detection of a lower neutron fluence.
(c) The possibility of using the same detector for measuring the gamma dose and the neutron dose results in a simplification of the personnel dosimeter.

Hence, investigations were made to determine (a) in what circumstances the Cerenkov effect can be utilized to measure the beta radiation induced by neutrons, (b) what is the detection sensitivity obtained for neutrons, (c) what are the advantages against conventional methods of measurement, and (d) what practical possibilities of application seem to be advantageous for health physics purposes.

First, the investigations were restricted to activation detectors for detecting fast neutrons by the threshold reactions ^{31}P (n, p)^{31}Si and ^{32}S (n, p)^{32}P (see Table I). Compounds containing sulphur and phosphorus in a liquid form were used as well as silver-activated metaphosphate glasses as solid-state detectors. The Cerenkov radiation was directly measured in an assembly of the type used in the liquid scintillator method. The activation sample was placed between two photomultipliers whose coincidence counting rate was a measure of the beta activity.

TABLE I. CHARACTERISTICS OF P AND S PROBES FOR THE DETECTION OF FAST NEUTRONS

	$^{31}P(n, p)^{31}Si$	$^{32}S(n, p)^{32}P$
Threshold energy	2.7 MeV	2.8 MeV
Half-life	2.6 h	14.2 d
Emission per decay	1.5 MeV β^-, 100%	1.7 MeV β^-, 100%
Effective efficiency:		
Fission neutrons	34 mbarn	65 mbarn
Pu-Be neutrons	90 mbarn	230 mbarn
14-MeV neutrons	85 mbarn	245 mbarn

2. METHOD OF MEASUREMENT

2.1. Theory

In a transparent medium, charged particles can attain a speed higher than the speed of light in the same medium. If $n \times \beta$ is larger than 1 (n = refractive index, $\beta = v/c$ relative particle velocity in the medium) polarized light is emitted in the visible range of light wavelengths. The Cerenkov effect is used, in particular, to identify high-energy particles in particle accelerators which strike a detector under a certain direction of incidence, and to discriminate them against other background radiation [5].

The possibility of using the Cerenkov effect for the detection of gross beta radioactivity of water samples for health physics purposes was also investigated. It was shown that beta particles with energies above 1 MeV can be detected by the Cerenkov effect in water. This method of measuring can be used mainly for higher activity concentrations after accidents to measure fission-product mixtures of a known composition or other defined radionuclides [6, 7]. Since emitters with energies below 1 MeV are not measured, a transfer of the method to the measurement of neutron activation detectors offers the advantage that the beta activity of ^{32}P (E_{max} = 1.7 MeV) and ^{31}Si (E_{max} = 1.5 MeV) can be determined without any influence by spurious low-energy radiation components.

2.2. Description of the measuring set-up

For the measurement of the Cérenkov effect in liquids and solids, commercial equipment for liquid scintillator measurements (tritium measurements in an aqueous solution) by Packard[1] and Beckman was used. The sample was placed between two photomultipliers and the coincidence count-rate measured. Even in the measurement with glass dosimeters, the usual plastic bottles of about 20-ml volume were used with special plexiglas inserts which retained the glass in the centre of the sample bottle.

With a glass cube 8 mm × 8 mm × 4.7, the measuring effect due to the Cerenkov radiation can be enhanced by detection of the beta particles emitted from the glass in the surrounding medium. Among the various possibilities of measurement, i.e. glass in air, glass in water, and glass in plexiglas, plexiglas was selected for its favourable handling (see Table II). The background of the measuring set-up determines the lower detection limit of the method of measurement. Its measurement for each set-up was made with an inactivated sample. The size of the inactivated

TABLE II. DETECTION SENSITIVITY OF YOKOTA GLASS BY CERENKOV MEASUREMENT IN THE LIQUID SCINTILLATION COUNTER

Method of measurement	Counting rate (counts/min)	Rel. counting rate (%)
Glass in air	1105	89
Glass in plexiglas	1244	100
Glass in water	1480	119
Glass in plexiglas and water	1493	120

1 For measurements of liquid activation samples only.

sample depends on the type of measurement sample and the equipment used; in the samples studied here the background count-rate was between 16 and 25 counts/min.

2.3. Discriminator setting for Cerenkov measurement

Corresponding to the different maximum beta energies of the radionuclides, different discriminator values have to be set for the measurements of ^{32}P, ^{14}C or ^{3}H samples. Therefore, pulse-height spectra were recorded with a constant window aperture of 2% and a variable discriminator setting for the various radionuclides. The pulse-height spectra of Cerenkov radiation, which were produced by ^{32}P in solution and in the phosphate glass, respectively, are shown in Figs. 1 and 2 compared with liquid scintillator spectra of ^{32}P, ^{14}C and ^{3}H. The light generated in an aqueous solution or in a transparent solid accordingly shows roughly the same pulse-height distribution as the radionuclide tritium in a liquid scintillator.

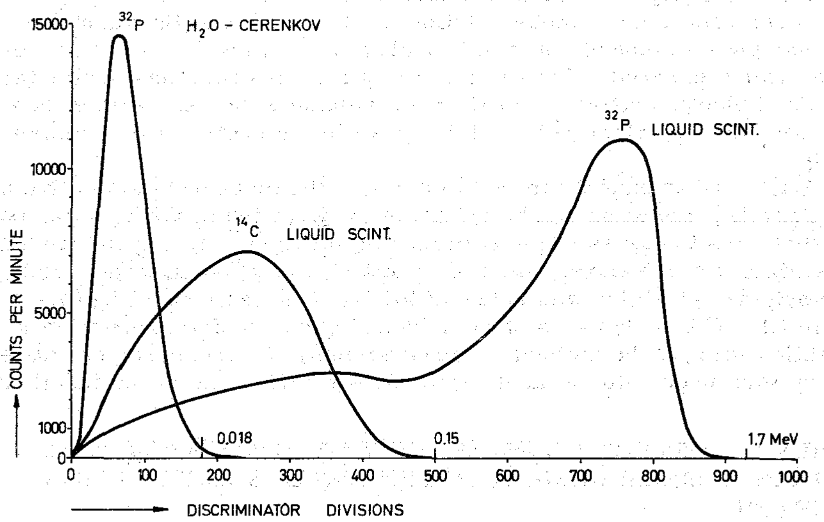

FIG. 1. Pulse-height spectrum of ^{32}P and ^{14}C in liquid scintillator solution compared with ^{32}P in aqueous solution recorded with the Beckman liquid scintillation counter

Colouration of the sample results in a shift of the pulse-height distribution towards smaller pulses and thus in a reduction in the integrated counting rate because of the absorption of light in the measurement of the Cerenkov effect, just as in the measurement in a scintillator solution. However, a colouration of the phosphate glass by additional gamma doses effective in this way was only found above a dose of 1000 R. This colouration at higher doses changes the spectrum of the Cerenkov measurement and the amount of the counting rate in the way shown in Fig. 3 for two glasses with the same neutron irradiation, but with different measured gamma values.

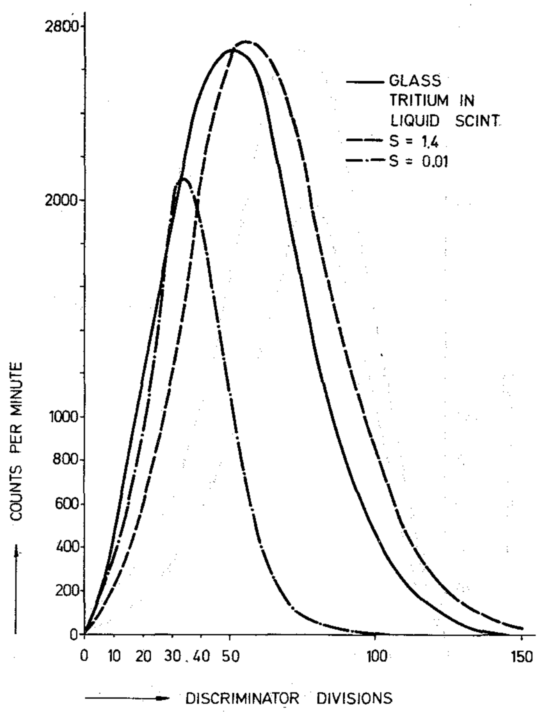

FIG. 2. Pulse-height spectrum of Cerenkov radiation of a neutron-activated phosphate glass compared with the pulse-height spectrum of a ^3H-containing liquid scintillation sample with different quenching factors, measured with the Beckman liquid scintillation counter

The measured values of the Cerenkov measurements quoted below correspond to the integral counting rate in the range of pulse heights normally used as the basis for tritium measurements in the set-up (discriminator setting 10 to 120).

2.4. Irradiation experiments

The activation samples were irradiated with neutrons of different energies and varying neutron fluence, namely:

With thermal neutrons in the thermal column of the reactors FR 2 and SUR (10^9 to 10^{12} n/cm^2).

With fast neutrons from a 10-Ci Pu-Be source (10^7 to 10^8 n/cm^2).

With 14.1-MeV neutrons from neutron generators by the (T, d) reaction (10^7 to 10^{10} n/cm^2).

The irradiation periods ranged from several minutes to several hours. The calibration for fast neutrons was made by measuring the beta radiation of ^{32}P in a sulphur pellet, and for thermal neutrons by measuring the beta radiation of ^{198}Au in a gold foil with a proportional flow counter. The measured values of the Cerenkov measurement were converted, as described in section 3, for the long-time irradiation necessary with Pu-Be neutrons to the maximum value of activity after a short-time irradiation.

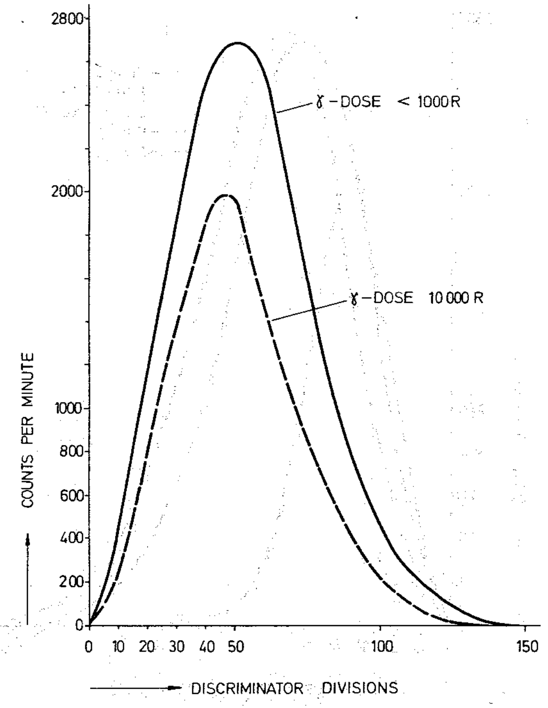

FIG. 3. The change of the pulse-height spectrum of Cerenkov radiation of a neutron-activated phosphate glass by colouration of the glass by gamma doses above 3000 R.

This was done to enable a direct comparison of the individual reactions for application in criticality dosimetry.

3. CERENKOV EFFECT IN LIQUID ACTIVATION SAMPLES

3.1. Dose sensitivity of various detectors

The Cerenkov measurement, e.g. in an activation detector available in an aqueous solution, offers the advantage of direct measurement of ^{32}P and ^{31}Si without the addition of a liquid scintillator and without enrichment. First, the sensitivity of detection attained for fast neutrons with the Cerenkov measurement should be ascertained. The activation detectors used were simple substances, namely: ammonium phosphate (3 g) in an aqueous solution; ammonium sulphate (3 g) in an aqueous solution; and carbon disulphide (20 g). These detectors were filled into the plastic bottle provided for liquid scintillator measurements (see also Ref. [8]).

Activation of the samples with Pu-Be neutrons resulted in the dose sensitivity shown in Fig. 4 referred to an FC-dose as well as to a short-time irradiation. The decrease in the relative counting rate is shown in Fig. 5 as a function of the time after exposure.

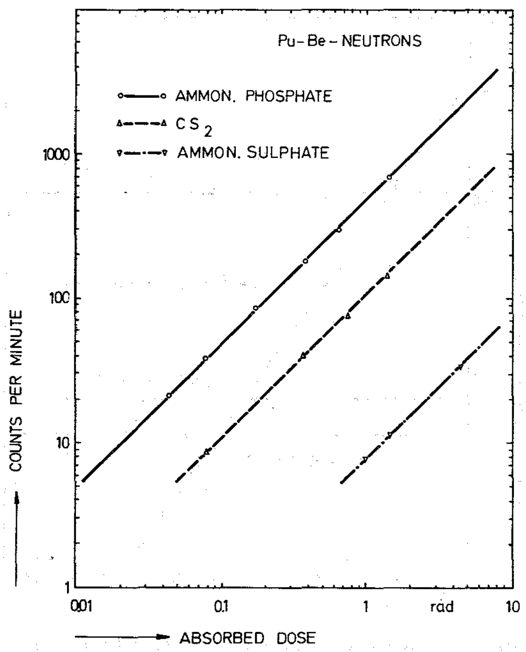

FIG. 4. The count-rate of Cerenkov radiation of different aqueous activation detectors as a function of the FC dose of fast neutrons (Pu-Be neutrons) measured with the Packard Tricarb
O——O Ammonium phosphate, 3 g in 20 ml H_2O
Δ- - -Δ Cs_2, 20 g
∇—·—∇ Ammonium sulphate, 3 g in 20 ml H_2O

Because of its sulphur content, the ammonium sulphate solution showed a low dose sensitivity compared with the dose sensitivity of the carbon disulphide detector. The sensitivity of ammonium phosphate is the highest of the three detectors, but it has advantages only for short-time irradiation and immediate evaluation, since in routine daily evaluation of the activation detectors there may be 6-8 h between exposure and measurement in the most unfavourable case.

3.2. Detection limit of various measuring methods

The limit of detection is set by the background, the specific dose sensitivity of the detector and the half-life of the radionuclide to be measured, hence by the time of the measurement.

The limit of detection obtained with Cerenkov measurement with various liquid activation samples was compared with the limit of detection obtained with a sulphur pellet. For this purpose the sulphur pellet was counted directly or after incineration in a 5-cm-diam. dish in a proportional flow counter with a 1 mg/cm^2 window foil (protective counter with anticoincidence and 5 cm of lead shielding).

Table III gives the values of background, relative dose sensitivity, and calculated efficiency for the different activation samples. Because of the low background, when measuring with a proportional counter, a

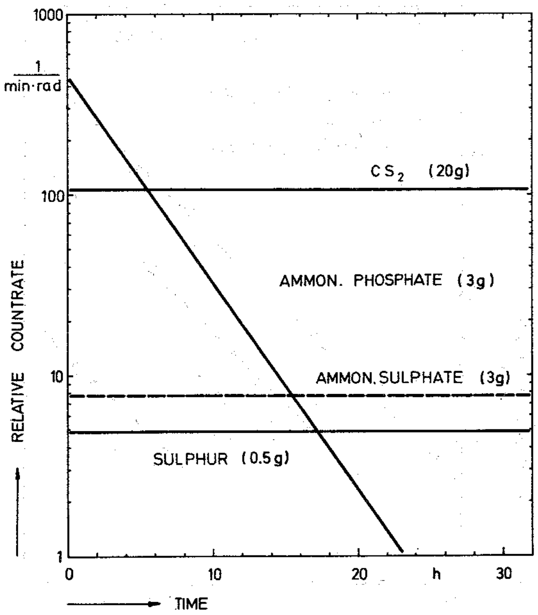

FIG. 5. The relative pulse-rate of activation substances related to an FC dose of Pu-Be neutrons as a function of the time after irradiation. Measurement of the Cerenkov radiation in the Tricarb.
Ammonium phosphate, 3 g in 20 ml of aqueous solution after short-time irradiation
Carbon disulphide, 20 g
Ammonium sulphate, 3 g in 20 ml of aqueous solution
Sulphur pellet (0.5 g) burned up, measurement of the beta activity in the proportional flow counter
(Here, 1 rad corresponds to a neutron fluence of 2.56×10^8 n/cm^2)

sulphur pellet is better than liquid activation samples for the detection of low doses.

The measurement of ^{32}P in an aqueous solution with a liquid scintillator would result in 95% efficiency. However, since only 2 ml of phosphorous solution can be used in the 20-ml sample, this only results in an effective efficiency of 9.5%, which must be compared with the ammonium phosphate solution efficiency of 30.6%.

Hence, the determination of beta activity of liquid activation probes by Cerenkov radiation has advantages only when the size of the activation detector is not prescribed (area dose), when incineration, evaporation, chemical separation, or coprecipitation are impossible for lack of time or because of potential losses, and when it is possible to measure in the liquid phase during the chemical preparation of low activities.

4. CERENKOV EFFECT IN SILVER-ACTIVATED METAPHOSPHATE GLASSES

4.1. Method of detection

Silver-activated metaphosphate glass, in particular sensitive Low-Z glass with a low pre-dose, is increasingly used as a gamma dosimeter in routine dosimetry. Because of the type of additives in glass and the use of basic material of very high chemical purity in glass manufacture, glass can also be regarded as an ideal activation detector (see Table IV). Thus, phosphate glass permits simultaneous dose measurement after a criticality accident of gamma radiation and neutrons. The possibility of detecting both types of radiation with only one detector would make it unnecessary to carry additional activation detectors.

Therefore, an investigation was made into the possibility of detecting neutrons by the induced activity of substances inherent in the glass, mainly by the reaction ^{31}P (n, p) ^{31}Si for fast neutrons and by the reaction ^{31}P (n,γ) ^{32}P for slow neutrons, by means of direct measurement of the Cerenkov radiation generated in the Yokota glass. The different half-lives of the radionuclides ^{31}Si and ^{32}P (2.6 h and 14.2 d respectively) and the different activation cross-sections for thermal and fast neutrons may, under certain conditions, allow the separate detection of thermal and fast neutrons, despite other spurious activities with different half-lives originating at the time of irradiation.

4.2. Neutron detection in glass

In the measurement of the fluorescence caused by (n, α) processes and by the capture of slow neutrons in lithium, boron, and silver, the phosphate glass is sensitive particularly to thermal neutrons. The sensitivity to thermal neutrons may be adapted to the gamma-dose sensitivity (total dose-equivalent of thermal neutrons and gamma radiation) in routine dosimeters by a special spherical capsule containing boron [10, 11]. It is not possible to detect fast neutrons by fluorescence measurements because of the low sensitivity, as the neutron-induced beta and gamma radionuclides with half-lives of 24 s to 253 d (see Tables V and VI) do not make any significant contribution to the formation of radiophotoluminescence centres [11, 12].

It is true that the Cerenkov radiation permits the detection of fast neutrons by the radionuclides ^{31}Si and ^{32}P, but the measurement may be disturbed immediately after irradiation by neutron-induced short-lived emitters with high-energy beta radiation above 0.5 MeV. The change in the counting rate with time of the Cerenkov radiation was experimentally determined immediately after a short-time irradiation with 14 MeV and thermal neutrons (Figs. 6 and 7). Already 1 h after short-time irradiation only the ^{31}Si and ^{32}P, fractions of the measured value were detected, but no other spurious components. The decrease in the counting rate with time after a while showed the characteristic half-lives of the two radionuclides to be 2.6 h and 14.2 d, respectively. Hence, phosphate glass is suitable for use as an activation detector for the detection of thermal neutrons as well as fast neutrons.

4.3. Fluence sensitivity to fast and thermal neutrons

To determine the detection sensitivity of the glass to fast neutrons, phosphate glass was irradiated at varying neutron fluences. Figure 8 shows the counting rate of Cerenkov radiation as a function of neutron fluence for 14 MeV and Pu-Be neutrons. The measured value of unirradiated glass is 20 counts/min.

TABLE III. THE DETECTION LIMIT OF FAST NEUTRONS BY THE MEASUREMENT OF CERENKOV RADIATION OF ACTIVATING SUBSTANCES CONTAINING EITHER P OR S

Detector	Quantity	Method of measurement	Background (counts/min)	FC-dose sensitivity		Efficiency (%)	FC-dose (rad) with back-ground doubl.
				Counts/min per rad	Counts/min per g × rad		
Ammonium phosphate	3 g in 20 ml of aqueous solution	Cerenkov radiation in the Tricarb	22.9	465	655	30.6	0.035
Ammonium sulphate	3 g in 20 ml of aqueous solution		23.4	7.7	10.0	32	2
Carbon disulphide	20 g liquid		16.7	108	6.75	17.5	2
Sulphur	About 0.5 g solid	Proportional flow counter (anti-coincidence)	4.6	2.1	4.0	13	1
Sulphur	About 0.5 g burned up		4.6	4.9	9.4	30	0.5

TABLE IV. COMPOSITION OF SILVER-ACTIVATED METAPHOSPHATE GLASS ACCORDING TO YOKOTA [9]

Element	Relative weight per cent
Li	3.6
B	0.85
O	53.7
Al	4.6
P	33.5
Ag	3.7

A neutron fluence of about 5×10^7 n/cm^2, referred to short-time irradiation, accordingly results in doubling the background for fast neutrons above 2.5 MeV. For fission neutrons the value is about three times higher. The detection of fast neutrons can be falsified by thermal neutrons produced simultaneously by the neutron capture reaction $^{31}P\ (n, \gamma)\ ^{32}P$. Since phosphate glasses are worn in capsules in routine dosimetry to compensate for the energy dependence on gamma radiation, the counting rate of the Cerenkov measurement for glass in various capsules was determined as a function of the neutron fluence of thermal neutrons for a Yokota glass without any capsule, for a glass in a spherical capsule, and in a capsule of 1 mm of cadmium. The capsule of 1-mm cadmium absorbs all thermal neutrons. The spherical capsule containing boron reduces the activity fraction in the glass to some 20% compared with the non-encapsulated glass. A neutron fluence of about 1.7×10^{10} n/cm^2 of thermal neutrons results in doubling the background of the Cerenkov measurement in the spherical capsule.

4.4. Separation of thermal and fast neutrons

A separation of the fractions of the measured values into thermal and fast neutrons is often possible. The activation fraction of fast neutrons decays with a half-life of 2.6 h. In this way one can determine the activation fraction of thermal neutrons exclusively by another measurement about 10-20 h after exposure and deduct it from the measured total. Figure 10 shows the example of a reactor irradiation, pointing out the decrease of the counting rate of the Cerenkov measurement after the end of irradiation for a non-encapsulated glass. The change in counting rate with time of the Cerenkov measurement for a phosphate glass in a spherical capsule is shown in Fig. 11. The dosimeter was irradiated at the same fluence of thermal and fast neutrons. A higher fraction of thermal neutrons has not been observed so far in any criticality accident. For the measurement of fast neutrons, the sensitivity for fission neutrons was taken as the basis. Within the first 10 h after a criticality accident a correction in the determination of the activation percentage of fast neutrons

TABLE V. DETECTION OF THERMAL NEUTRONS IN THE YOKOTA GLASS

Nuclear reaction	Isotope fraction (%)	Half-life	Cross-section (barn)	Emission per decay	
				β	γ
$^{31}P(n,\gamma)\,^{32}P$	100	14.3 d	0.191	1.71 MeV β^-, 100%	-
$^{107}Ag(n,\gamma)\,^{108}Ag$	51	2.4 min	44	1.65 MeV β^-, 95% 1.02 MeV β^-, 1.7% 0.88 MeV β^-, 0.28%	0.63, 0.43 MeV 2.2%
$^{109}Ag(n,\gamma)\,^{110}Ag^m$	49	253 d	2.8	0.085 MeV β^-, 62.5% 0.53 MeV β^-, 36.9% 1.5 MeV β^-, 0.6%	0.66, 0.89, 0.94 1.38, 0.76, 0.71 MeV (100%)
$^{109}Ag(n,\gamma)\,^{110}Ag$	49	24 s	110	2.86 MeV β^-, 58% 1 MeV 29% 2.2 MeV 12.5%	0.66 MeV, 100%
$^{27}Al(n,\gamma)\,^{28}Al$	100	2.3 min	0.21	2.87 MeV β^-, 100%	1.78 MeV, 100%

TABLE VI. DETECTION OF FAST NEUTRONS IN THE YOKOTA GLASS

Nuclear reaction	Isotope fraction (%)	Half-life	Threshold energy (MeV)	Emission per decay	
				β	γ
^{31}P (n, p) ^{31}Si	100	2.6 h	2.7	1.48 MeV β^-, 100%	1.26 MeV, 0.07%
^{31}P (n, 2n) ^{30}P	100	2.55 min	12.3	3.24 MeV β^+, 99%	2.24 MeV, 0.5%
^{31}P (n,α) ^{28}Al	100	2.3 min		2.87 MeV β^-, 100%	1.78 MeV, 100%
^{27}Al (n, p) ^{27}Mg	100	9.45 min	4.7	1.75 MeV β^-, 58% 1.59 MeV β^-, 42%	1 MeV, 30% 0.84 MeV, 70%
^{27}Al (n,α) ^{24}Na	100	15 h	7.3	1.39 MeV β^-, 100% 4.14 MeV β^-, 0.003%	1.37 MeV, 100% 2.75 MeV, 100%
^{107}Ag (n, 2n) ^{106}Ag	51	24.5 min	9.6		0.51 MeV, 126% 0.19 MeV, 100%
^{109}Ag (n, p) $^{109}Pd^m$	49	4.75 min			0.305 MeV, 0.07%
^{109}Pd	49	13.5 h		1.03 MeV β^-, 100%	
^{109}Ag (n, 2n) ^{108}Ag	49	2.4 min		1.65 MeV β^-, 95% 1.02 MeV β^-, 1.7% 0.88 MeV β^+, 0.28%	0.63 MeV 0.43 MeV
^{16}O (n, 2n) ^{15}O		2.05 min	16.6	1.73 MeV β^+	(0.51 MeV)

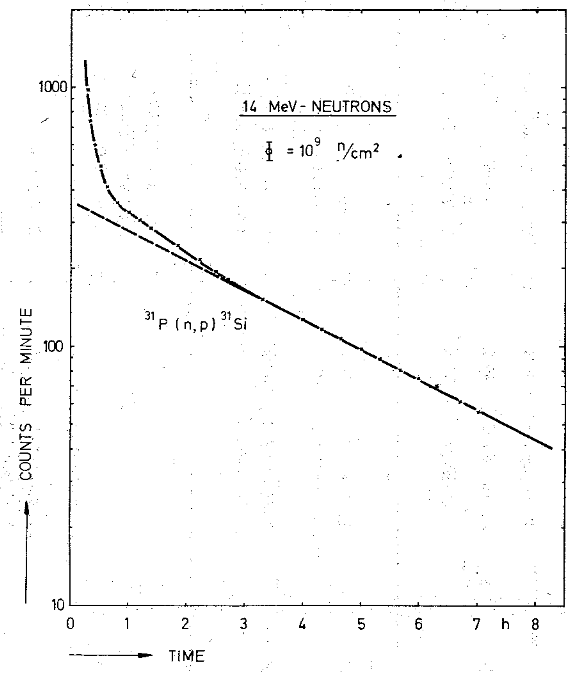

FIG. 6. The count-rate of Cerenkov radiation of a Yokota glass as a function of the time after exposure. Short-time irradiation with 14-MeV neutrons

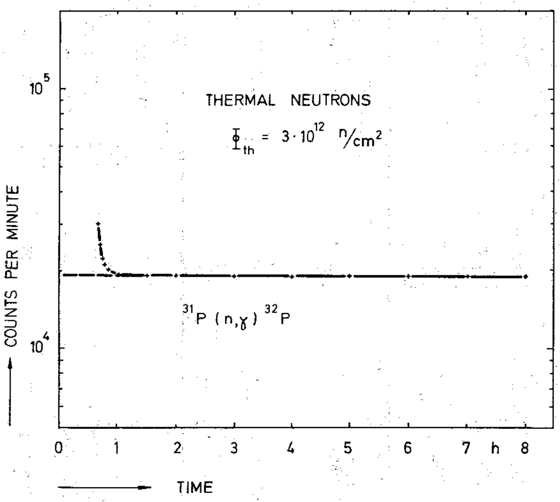

FIG. 7. The count-rate of Cerenkov radiation of a Yokota glass as a function of the time after exposure. Short-time irradiation in the thermal column of the reactor FR 2

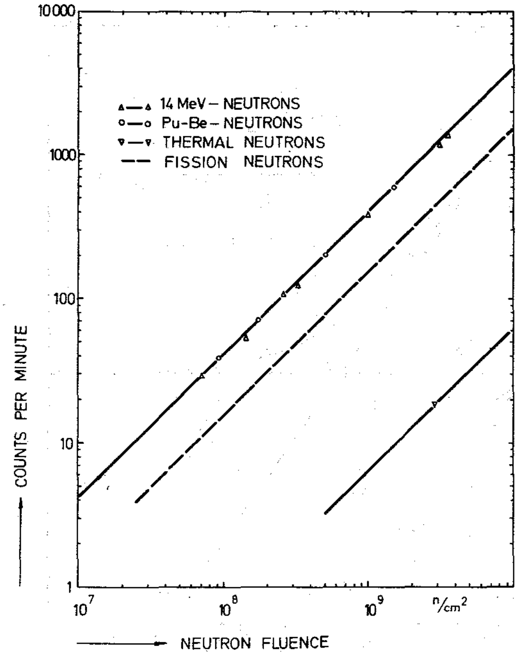

FIG. 8. The count-rate of Cerenkov radiation of a Yokota glass as a function of the neutron fluence for: 14-MeV neutrons; Pu-Be neutrons; thermal neutrons

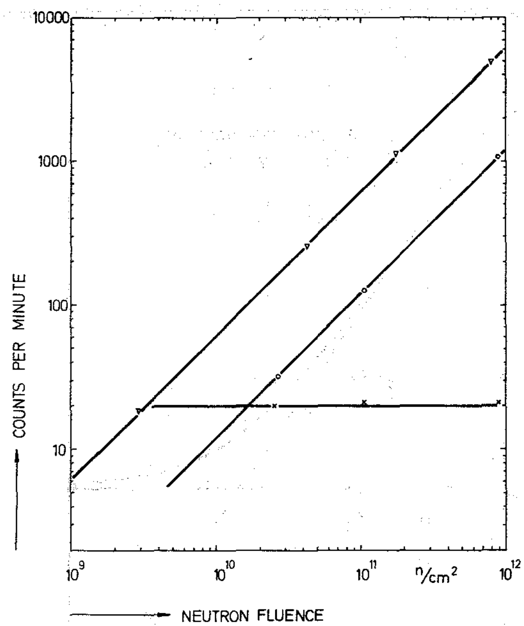

FIG. 9. The count-rate of Cerenkov radiation of different phosphate glass dosimeters as a function of the fluence of thermal neutrons.

▽——▽ Yokota glass without capsule

○——○ Yokota glass within a boron-containing capsule

×——× Yokota glass in a capsule of 1-mm Cd

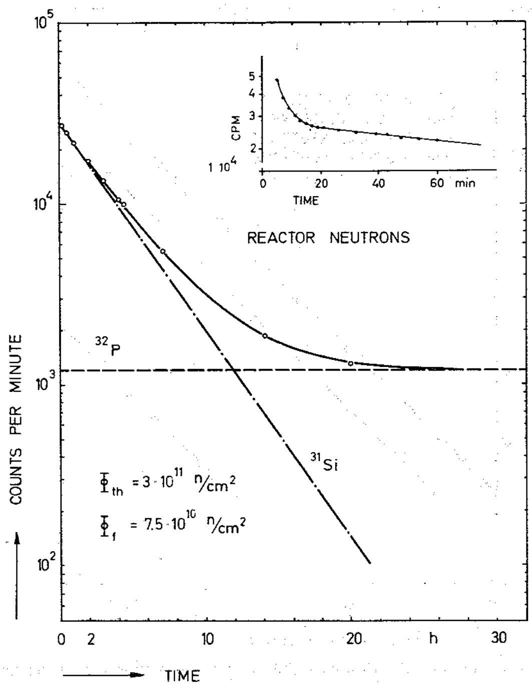

FIG. 10. The count-rate of Cerenkov radiation of a non-encapsulated Yokota glass as a function of the time after exposure. Neutron activation in the reactor core of the uniform moderated zero-energy reactor SUR

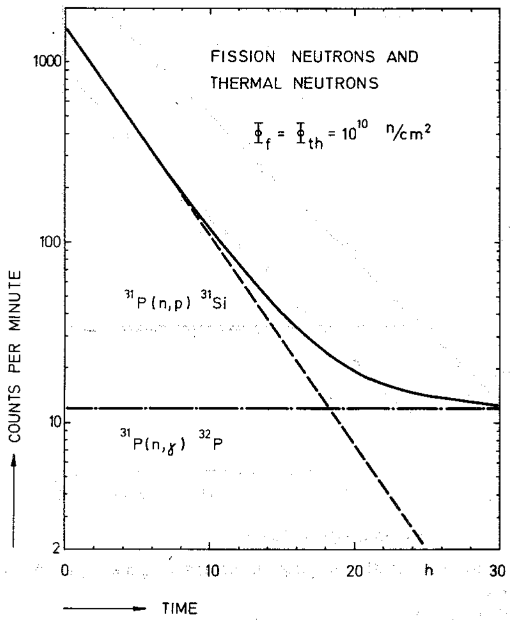

FIG. 11. The count-rate of Cerenkov radiation of a Yokota glass in a spherical capsule as a function of the time after exposure. Neutron activation with 10^{10} n/cm² of thermal and fission neutrons

TABLE VII. DETECTION OF BETA ACTIVITY IN THE YOKOTA GLASS (8 mm × 8 mm × 4.7 mm)

Method of measurement	Background (counts/min)	Counting rate (counts/min)	Rel. counting rate (%)
Cerenkov measurement			
In plexiglas	20	1244	100
In H_2O		1480	119
Proportional counter			
(a) Inside measurement			
On Al[a]	65	494	40
On Pb[a]	84	509	41
(b) Outside measurement I			
On Al[a]	38	362	29
On Pb[a]		390	31
(c) Outside measurement II			
On Al[a]	11	234	19
On Pb[a]		290	23

[a] Backscatter foil.
Measurements I and II mean measurements with differently constructed proportional counters with 1 mg/cm^2 window foil.

for the thermal activation percentage is not necessary. It can be determined by another or later measurement of the glass, or perhaps by repeated measurements.

Owing to the relatively short half-life of the produced radionuclide ^{31}Si and the rise in the detection limit, phosphate glass can be used as an activation detector only for criticality dosimetry where the time of irradiation is known and measurement can be made shortly after.

4.5. Detection limit for other methods of measurement

To obtain a comparison with the Cerenkov measurement, the beta activity of phosphate glass was also counted with proportional counters. Methane flow counters of various types were used. The phosphate glass

TABLE VIII. METHODS OF DETECTION IN NEUTRON DOSIMETRY

Method	Detector	Energy range	Neutron detection (fluence detection)	Storage of measured value
Nuclear-track registration				
Photographic emulsion	Nuclear-track emulsion	(n_{th}), $n_f > 0.5$ MeV	$> 5 \times 10^5$ n/cm^2	~ 30 d
Glasses with fission foil	Glass + foil of fissile material (e.g. ^{238}U)	(n_{th}), $n_f > 2.0$ MeV	$> 2 \times 10^7$ n/cm^2 [a]	(Unlimited)
		n_{th}	$> 2 \times 10^7$ n/cm^2	
Activation measurement				
Beta, gamma activity	S	$n_f > 2.5$ MeV	$> 5 \times 10^7$ n/cm^2	$T_H = 14.2$ d
Cerenkov radiation	Glass	$n_f > 2.5$ MeV	$> 2 \times 10^7$ n/cm^2 [b]	$T_H = 2.6$ h
		n_{th}	$> 5 \times 10^9$ n/cm^2	$T_H = 14.2$ d
Photoluminescence measurement				
Thermoluminescence dosimeter	LiF	γ, n_{th}	$> 10^5$ n/cm^2	(Unlimited)
Phosphate glass dosimeter	Phosphate glass	γ, n_{th}	$> 10^7$ n/cm^2	
Change in solids	Silicon diodes	$n_f > 0.3$ MeV	$> 2 \times 10^8$ n/cm^2	(Unlimited)

[a] The detection limit is given by the background (spontaneous fissions).
[b] 1 hour after short-time irradiation.

was measured outside the counting chamber behind a foil of 1 mg/cm^2 and inside the counting chamber. The results of the comparison are listed in Table VII. The good efficiency of the inside counter is compensated for in practical use by the relatively high background of the set-up and by the required scavenging time of some 10 min. The Cerenkov measurement of phosphate glass in comparison with the other sensitive methods of detection offers the advantages of a short total time for evaluation and a low limit of detection. In this set-up also measurements of more than 10^6 counts/min are possible without dead-time correction due to the fast coincidence stage.

To determine the absolute efficiency of various detectors, the detection probability of phosphate glass to neutrons was calculated on the basis of the size of the glass cube, the respective cross-section, neutron fluence, and the decay constant. For the detection of beta radiation by means of the Cerenkov radiation produced in the glass, an efficiency of about 60 ± 5%, with the errors in measurement taken into account, was obtained from the Beckman instrument used under optimum conditions (glass in plexiglas and water, see Table II).

Table VIII shows a compilation of the properties of various methods of measurement which may be used in personnel dosimetry for the detection of neutrons. The measurement of fast neutrons by Cerenkov radiation in a phosphate glass dosimeter thus shows a limit of detection comparable with that of other accident dosimeters, but a more unfavourable long-time behaviour.

5. THE USE OF PHOSPHATE GLASS AS A CRITICALITY DOSIMETER

A silver-activated phosphate glass – originally intended only for the measurement of a gamma dose – can be used as a criticality dosimeter for the detection of various components of radiation:

The fluorescence measurement of the glass permits the determination of the gamma dose.

The measurement of the Cerenkov effect of beta radiation of ^{31}Si permits the determination of the dose of fast neutrons above 2.5 MeV by the nuclear reaction $^{31}P\,(n, p)\,^{31}Si$.

The measurement of the Cerenkov effect of beta radiation of ^{32}P permits the determination of the thermal neutron dose by means of the nuclear reaction $^{31}P\,(n, \gamma)\,^{32}P$.

Besides, it should be pointed out that the analysis of induced gamma activity can give some information about the time of irradiation.

In the following sections a routine dosimeter for gamma radiation (Yokota glass 8 mm × 8 mm × 4.7 mm in a spherical capsule) is used to show the lower limit of detection to be expected for neutrons in a criticality accident.

In the first few hours after the cricicality accident a measured value is obtained by the Cerenkov radiation for the total dose of fission neutrons and thermal neutrons. The measured value and thus the dose sensitivity for fast neutrons decrease rapidly because of the half-life of ^{31}Si (Fig. 12), whereas the measured value for thermal neutrons remains practically the same.

The lower dose detection limit for fast neutrons is thus determined essentially by the time of measurement after the accident and by the

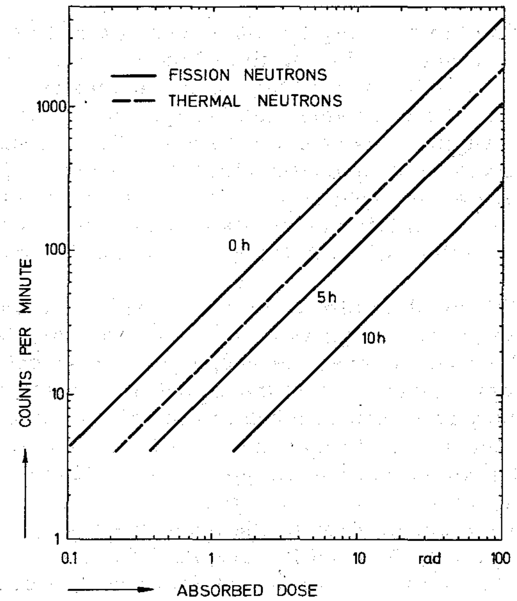

FIG. 12. The dose sensitivity for fission neutrons and thermal neutrons received by Cerenkov measurements of a Yokota glass in a spherical capsule at varying times after a short-time irradiation. (1 rad of a surface dose generated by heavy, charged particles corresponds to a neutron fluence of 2.76×10^8 n/cm^2 of fission neutrons and 1.59×10^{10} n/cm^2 of thermal neutrons.)

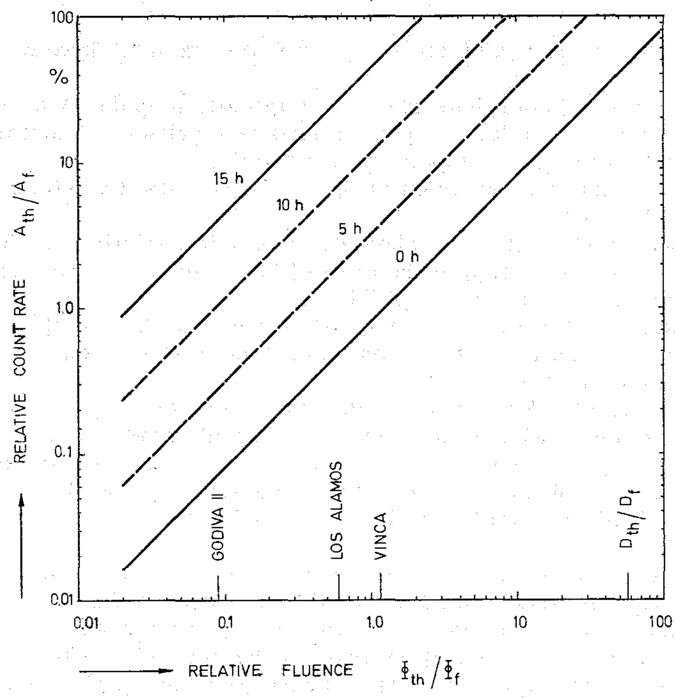

FIG. 13. The relative count-rate fraction for thermal neutrons as a function of the relative fluence fraction of thermal neutrons, relative to the fission neutron component of a neutron spectrum. Measurement of Cerenkov radiation at different times of measurement after a short-time irradiation

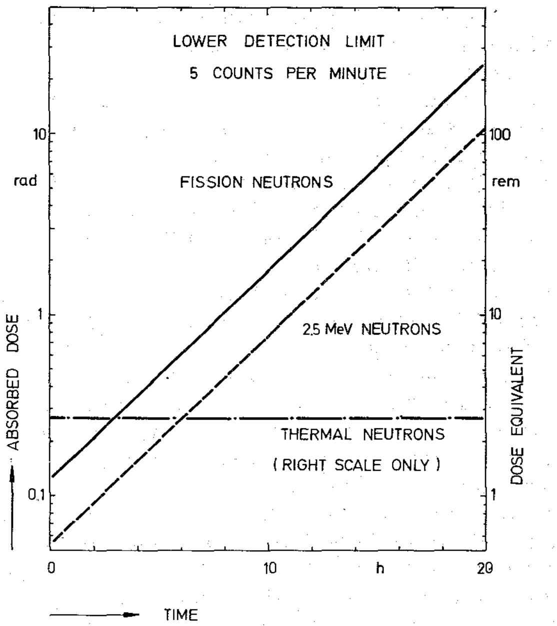

FIG. 14. The lower detection limit (5 counts/min) of Cerenkov radiation as a function of the time of measurement after a short-time irradiation for thermal and fast neutrons with a Yokota glass in a spherical capsule

amount of the thermal neutron fraction in the respective neutron spectrum. Figure 13 shows the increase in the relative counting-rate fraction of thermal neutrons as a function of the relative fluence fraction of thermal neutrons of a neutron spectrum, if the Cerenkov radiation measurement is carried out 5, 10, and 15 h after exposure. The figure shows the approximated fluence ratios Φ_{th}/Φ_f which were determined from the values measured by threshold detectors in familiar criticality accidents. Hence, for a criticality accident one can always assume a ratio less than 1. This means that the determination and potential separation of an activation fraction of fast neutrons by a routine dosimeter by Cerenkov measurement is possible within the first 15 h after short-time irradiation. This requires knowledge of the time of exposure. The smallest detectable dose of fast neutrons in a measurement 5 h after exposure is about 0.5 rads (for fission neutrons 1 rad of a surface dose generated by heavy, charged particles corresponds to 2.76×10^8 n/cm^2) (see Fig. 14).

Similar values are produced by phosphate glass of a different glass composition if there are no additional spurious activities. In other capsules the changed neutron sensitivity of the set-up must be taken into account.

6. APPLICATIONS

One example of the use of the Cerenkov effect for the determination of neutron-induced activities was shown by the nuclear reactions $^{31}P(n, p)^{31}Si$ and $^{32}S(n, p)^{32}P$ in liquid and solid-state detectors. Compared with other methods of measuring beta activity, the direct measurement of Cerenkov radiation in the solid offers a shorter total time for evaluation and a lower limit of detection.

The example of a phosphate glass dosimeter was given to show that this type of routine dosimeter for gamma radiation can also be used as an accident dosimeter under the conditions of a criticality accident for the determination of neutrons in a mixed radiation field. Here, the measurement of the Cerenkov effect in the phosphate glass permits a separation of the dose fractions contributed by fast and thermal neutrons within the first 15 h after a criticality accident.

For routine dosimetry of fast neutrons there is the possibility of obtaining an improved dose sensitivity by increasing the detector substance. A combination of solids containing phosphorus and sulphur might therefore be used in the design of a pocket-type dosimeter as a sensitive short-time routine dosimeter as well as a long-time dosimeter for determining a 14-d dose. Moreover, a dosimeter of this type would indicate thermal neutrons in a dose-correct way and could be used as a gamma dosimeter as well by fluorescence measurement.

Other applications of Cerenkov measurement in solid and plastic detectors were found in the detection of both high-energy neutrons and high-energy particles in the environment of particle accelerators. Neutrons above 20 MeV can, for instance, be detected by the nuclear reactions $^{12}C(n, 2n)^{11}C$ in a plastic detector without scintillator addition by means of the beta$^+$ radiation of ^{11}C.

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DISCUSSION

D. L. BLANC: With water the Cerenkov effect has an energy threshold at about 1 MeV for β-rays. What percentage of the β-rays emitted can you detect? I should like to ask a similar question for the case of glass, which has a higher energy threshold.

E. PIESCH: The lowest β-particle energy one can detect in phosphate glass by the Cerenkov effect is about 1 MeV, as with the detection of β-particles in the aqueous solution. To calculate the absolute efficiency we have taken into account the content of radionuclide in the glass, the reaction cross-section, the measured neutron fluence and the decay constant. We found an efficiency of some 60% under optimum conditions.

D.L. BLANC: The light rays emitted by the Cerenkov effect are directional. Does this not result in a directional effect with the instrument you have designed?

E. PIESCH: The Cerenkov effect measurement with glass shows a directional dependence of about 20%, given by the glass geometry. The glass was therefore kept in the centre of the sample bottle with a fixed orientation.

D.L. BLANC: I should just like to add that in our laboratory we are making active-cathode GM counters for neutron dosimetry. I think their sensitivity is better, at least for 14-MeV neutrons, because we can count practically all the β-rays emitted in the sensitive volume with a geometry of approx. 2π steradians.

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