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Nuclear Accident Dosimetrv Measurements at the Secend I.A.E.Ä. Intercomparison at Oak Ridge, USA, May 1971

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Nuclear Accident Dosimetry Measurements at the Second I.A.E.A. Intercomparison at

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Abstract

Teams from several countries participated in an International Intercomparison of Nuclear Accident Dosimeter Systems, organized by the I.A.E.A., in which criticality dosimeters were exposed to mixed pulses of neutron and gamma radiation from the Health Physics Research Reactor at Oak Ridge. This report gives the final results of the measurements made with the personnel dosimeter system of the Karlsruhe Nuclear Research Centre.

The dose assessment both in free air and on phantom are based on standard interpretation of the routine personnel dosimeter, of the phosphate glass dosimeter and cf threshold detectors. During the experiment the dosimetrie properties of a belt of polycarbonate foil were studied using the neutron-induced recoils and (n,a) reaction in the plastic foil for a detection of neutrons > 1 MeV. The effect of fission neutrons on the reading of gamma dosimeters were studied on different phosphate glass dosimeters and LiF dosimeters.

Zusammenfassung

Arbeitsgruppen von verschiedenen Ländern nahmen an einem von der I.A.E.A. organisierten internationalen Vergleich von Unfalldosimetersystemen teil, bei welchem Kritikalitätsdosimeter am Health Physics Research Reactor in Oak Ridge mit einem gemischten Neutronen- und Gammastrahlungsblitz bestrahlt wurden. Der vorliegende Bericht gibt die abschließenden Meßergebnisse wieder, die mit dem im Kernforschungszentrum Karlsruhe benutzten Personendosimetersystem erhalten wurden.

Die Bestimmung der Dosis für die Freiluft- und Phantombestrahlungen erfolgte unter Zugrundelegung der Standardauswertemethode mit dem Routinepersonendosimeter, dem Phosphatglasdosimeter und den Schwellwertdetektoren. Bei dem Bestrahlungsexperiment wurden die dosimetrischen Eigenschaften eines Gürtels aus Polykarbonatfolie untersucht, mit welchem Neutronen oberhalb einer Schwelle von 1 MeV über neutroneninduzierte Rückstoßkerne und (n, α) Reaktionen in der Plastikfolie nachgewiesen werden. Der Einfluß von Spaltneutronen auf die Anzeige von Gammadosimetern wurde an verschiedenen Phosphatglasdosimetern und LiF Dosimetern untersucht.

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dosimeter in the spherical capsule

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1. Introduction

After the First International Comparison on Nuclear Accident Dosimetry Systems at the French Atom Energy Centre at Valduc in 1970 (1) , the I.A.E.A. organized a second intercomparison at O.R.N.L., Oak Ridge. The intercomparison experiments consisted of the exposure of dosimeters to pulsed radiation from the Health Physics Research Reactor, the simultaneous measurement of the detectors, a dose assessment for each exposure position and a comparison of the first results of the participants.During the meeting the participants discussed the concepts employed in criticality dosimetry as weIl as developments of better dosimeter systems and evaluation techniques. The I.A.E.A. also coordinates work on the collection and unification of leakage spectra from criticality assemblies.

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The Karlsruhe Nuclear Research Centre took part in the international intercomparison at Oak Ridge with the routine personnel dosimeter and experimental dosimeters. The present experiments give us the opportunity to expose the Karlsruhe dosimeter to different energy spectra cf neutrons and to compare our own results with the resuits from the other participants.

This report gives the final results of the Karlsruhe measurements. At the time of writing the results of the other participants are not yet available.

2. Irradiations

The Health Physics Research Reactor (HPRR) at O.R.N.L. is a bare unmoderated reactor, the core of which is composed of uranium enriched to 93 % alloyed with molybdenium (2). The reactor produces radiation bursts of about 8×10^{-16} fissions per pulse. By using shieldings positioned on a circle of 2 m radius a modified fission spectrum was obtained.

The reactor was used in three different conditions:

- Pulse 1: reactor unshielded
- Pulse 2: reactor shielded by steel of 13 cm thickness
- Pulse 3: reactor shielded by lucite of 12 cm thickness

For each burst the dosimeters of the participants.were irradiated in a free-air geometry (free air irradiation) and on the front side of two Bomab man-like phantoms (phantom irradiation) filled with 0.9 % NaCl solution. The detectors were positioned on an arc at a distance of 3 metres from the reactor. Phantom A was placed facing the reactor, Phantom B was placed facing 90° from Phantom A.

3. Dosimeters

The Karlsruhe Nuclear Research Center participated at the intercomparison study with the routine personnel dosimeter as well as with experimental detectors. Our routinely used standard dosimeter consists of a phosphate glass dosimeter in a spherical capsule and of a combination of activation detectors in an additional plastic badge (3) (see Fig. 1). The activation detectors are a sulphur pellet, a bare gold foil, a gold foil covered by indium foils inside a cadmium shielding, and as an experimental detector also bare and cadmium-covered arsenic glasses. The phosphate glass is also used as a neutron activation detector via the reactions $\frac{31}{P(n,p)}$ Si for fast neutrons > 2.5 MeV and $31P(n, \gamma)^{32}$ for slow neutrons (4). The γ -Dosimeter is a Yokota glass of size 8 x 8 x 4.7 mm² inside a boron loaded plastic sphere and a perforated tin filter of 2 mm thickness.

layers of about detectors are in contact with Makrofol N foils. Polycarbonate foils The experimentally used threshold detector locket consists of neptunium, Makrofol E, thorium and uranium detectors which were electroplated in $40 \mu g/cm²$ on steel platelets of 1.2 cm diameter. The (Makrofol E) were also used as threshold detectors to measure neutrons above 1 MeV via recoils and (n,a) reactions. A plastic belt of Makrofol E was placed about the trunk of phantom A.

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All dosimeters were exposed on the chests of phantom A and phantom B. In addition, LiF ribbons of size $3 \times 3 \times 1$ mm³ (Harshaw TLD 100, TLD 600 and TLD 700) and a new phosphate glass with low sensitivity to thermal neutrons (Schott RPL-V glass) were exposed.

4. Evaluation Method

4.1 Personnel_dosimeters

Two methods were used to assess the neutron dose:

- from the personnel criticality dosimeter a sulphur disc, a bare and a cadmium shielded gold foil were measured,
- from the phosphate glass the neutron-induced activation were measured From the phosphate glass the heutron-induced activation were measured
within the first 15 hours ($\frac{31}{3}$ si) and 24 hours ($\frac{32}{7}$) after the irradiation.

The neutron dose is calculated from the S and P activities on the basis of an unmoderated fission spectrum. The dose fractions of thermal and intermediate neutrons were determined from the Au - Au/Cd system or from ^{32}P assuming an intermediate $\frac{1}{E}$ spectrum and a thermal Maxwellian spectrum. For pulse 2 the fast neutron dose was corrected using assumptions about the shape of the spectrum based on neptunium measurements or on the activity ratio $31_{\text{Si}}/76$ As measured with the free air dosimeter.

4.2 Experimental_dosimeters

The threshold detector systemaonsists:of the sulphur disc in the routine dosimeter, and of the additional fissionable isotopes uranium-238, thorium-232 and neptunium-237, without using a boron shield. Neutroninduced recoils and (n,a) -Reactions in a Makrofol E foil serve also as a threshold reaction. The approximate threshold energies of 2.5 MeV, 1.5 MeV, 1.2 MeV, 1 MeV and 0.75 MeV were taken into account for sulphur, uranium-237, thorium-232, Makrofol E and neptunium-237 respectively. The $40 \mu g/cm^2$ thin oxyde layers of the fissionable materials are exposed in contact with Makrofol N foils. For the detection of fission fragment

tracks the foils were etched for 40 min in a 35 % KOH solution at 60°C, and for the detection of recoils and α -tracks for 4 hours in the same solution. The tracks were counted optically. The cross sections of the threshold detectors were estimated to rise abruptly at the threshold energy having a constant value above this threshold.

For the detection of intermediate neutrons a transparent As_2S_3g1ass (disc of 10 mm ϕ , 1 mm) and a phosphate-arsenic glass (8 x 8 x 4.7 mm³) were used bare and cadmium-covered. The reaction 75 As (n, $\gamma)$ 76 As $(T_H = 26$ hours) has a cross section curve similiar to that of sodium, but without resonance peaks, and with a higher value by a factor of 100 (5). In comparison to gold arsenic doesn't have the characteristics of a resonance detector. The activity ratio $31_{S1}/76_{As}$ of the arsenic glass measured with the free-air dosimeter gives an idea about the shape of the fission spectrum behind the reactor shielding.

Gamma-dose measurements were also performed with LiF-dosimeters. The corrections for the effect of neutron irradiation are nessessary for the sensitivity of $7L$ iF to fast, intermediate and thermal neutrons. A new silver-activated phosphate glass (RPL-V glass, from Schott u. Gen.), with a low sensitivity to thermal neutrons, was also used for the gamma dose measurement at the free-air stations. Here no neutron-absorber or correction were applied for the effect of neutrons.

5. Results

5.1 Neutron measurements

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For an intercomparison and interpretation of dose determination with that of other participants, the measured activities and fission rates of the neutron detectors have been converted to reaction rates normalised to 10¹⁰ atoms for each detector. The reaction rates for the neutron detectors in the personnel dosimeter exposed in free air are given in Tab. 1.

The results of fluence estimates are given in Tab. 2 and Tab. \bar{c} for the activation detectors gold, arsenic, phosphate glass in the boron loaded capsule, and for the threshold detectors respectively.

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The dose assessment for the personnel criticality dosimeter and the phosphate glass dosimeter are presented in Tab. 4.The results are given as Kerma (hectoerg \cdot g⁻¹). The gold foil and sulphur activities were used in the standard way to give the thermal, intermediate and fast neutron dose. A similiar evaluation was used for the activation reactions in the phosphate glass dosimeter. The results for fast neutrons were corrected for pulse 2 based on neptunium and arsenic measurements.

A summary of neutron dose estimates found with the routine dosimeter and/or the experimental detectors based on different detector combinations are presented in Tab. 5. The results for phantom B, which was arranged sideways to the reactor, are not consistent for the different detectors because of a shielding effect of the phantom and other dosimeters respectively.

The final results used for the comparison with those of other participants are presented in Tab. 6 and Tab. 7. The dose estimates are given in kerma (hectoerg \cdot g⁻¹) and absorbed dose in soft tissiue (rd). Free-air measurements were made with the personnel criticality dosimeter, the phosphate glass and the threshold detectors. The provisional results given during the Oak Ridge meeting are based on the phosphate glass dosimeter results. The gamma dose is corrected for the effect of slow neutrons.

The results for the personnel dosimeter exposed on the man-phantom are found with gold and sulphur detectors or with the phosphate glass activation. The absorbed doses at the front surface estimated with the standard interpretation were then converted to kerma using a constant conversion factor for the fission spectrum. The gamma dose given here is the total surface absorbed dose due to incident gamma rays and that produced by the $1_H(n, \gamma)^2$ D reaction in the phantom, which was measured with the phosphate glass dosimeter. The ²⁴Na activity of the phantoms were measured in a liquid scintillation counter. The sodium concentration of the phantom was 1.5 mg 25 Na/ml.

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ANGLE OF DETECTOR TO NEUTRON INCIDENCE

PERCENT OF NUCLEAR

5.2 Phantom studies with a belt of polycarbonate

A plastic belt of Makrofol E is a simple detector to determine the direction of the neutron incidence after a criticality accident. For testing the practical application of such a belt a 5 cm wide Makrofol strip welded into a thinner foil was exposed on the man-phantom. After irradiation discs of 1 cm diam were punched out of the belt at uniform distances, etched and counted microscopically. The relative number of recoil and (n, α) tracks is presented in Fig. 2 as a function of the position of the detector with respect to the angle of radiation incidence for the phantom A facing the reactor. The decrease of track number up to 90° is mainly due to the directional dependence of the detector foil. The effect of body or arm shielding, and of the energy threshold (1 MeV) on the dose estimation is a factor of $\overline{2}$ higher for the neutron spectrum moderated by 13 cm steel than for the other spectra. No difference in the number of tracks were found for all neutron spectra, if the plastic belt was exposed on a bottle phantom of 18 cm diam (5) . Also no fading effect was observed for the undeveloped etch pits in Makrofol E after one year. Phantom irradiations indicated that a remarkably strong effect is produced by the arms or ether scattering materials near the body, in particular for radiation incident at 90° (5).

5.3 Gamma dose estimation with phosphate glass and LiF dosimeters

The gamma dose presented in Tab. 6 and Tab. 7 were measured with the phosphate glass (Toshiba $FD-1$ glass) in the boron-loaded sphere. The correction for the effect of slow neutrons is based on the neutron induced $\bar{2}^2$ activity in the phosphate glass. A dosimeter reading of 10 rd inside the boron loaded sphere corresponds to 10 $^{\mathrm{10}}$ ⁿ/cm² (1).

The effect of thermal and intermediate neutrons on the glass reading were studied with two different filters (the routine dosimeter constists of a boron-loaded spherical capsule with perforated 2 mm tin and a capsule of 1.2 mm tin) and two glasses (Toshiba FD-1 and Schott RPL-V glass).

The gamma dose readings of the dosimeters is given in Tab. 8 for the free-air station. The boron-loaded capsule reduces the neutron effect on the dosimeter reading up to a factor of 2. The RPL-V glass is practically insensitive to thermal neutrons (1.2 rd per 10^{10} n/cm^2) (6).

The sensitivity of LiF dosimeters to thermal neutrons was studied on Harshaw TLD 100, TLD 600 and TLD 700 dosimeters (ribbons of size $3 \times 3 \times 1$ mm²) which were exposed as a dosimeter triple. Knowing the sensitivity of TLD 700, the difference of dosimeter readings TLD 600 -TLD 700 and TLD 100 - TLD 700 1s due to the effect of thermal neutrons. This value is independent of the gamma dose component and practically of the sensitivity to fast neutrons (Tab. 9). The results of these measurements are presented in Fig. \overline{z} as a function of the neutron fluence for TLD 600 and TLD 700. From this results average dosimeter readings of 600 rd. 213 rd and 1.3 rd were found corresponding to a fluence of 10¹⁰ n/cm² for TLD 600, TLD 100, and TLD 700 respectively.

In comparison to phosphate glasses TLD 700 has a remarkable sensitivity to fast and intermediate neutrons. For the assessment of gamma dose, corrections of the measured dose are necessary due to the effect of thermal, intermediate and fast neutrons. A calibration exposure with unmoderated fission neutrons in the field of a 252 Cf-neutrons source led to a correction of the dosimeter of about 1.25 rd per 10^{10} n/cm^2 for fast and intermediate neutrons (7). The response of TLD 700 to fast neutrons were found to be in good agreement with that from Tochilin whose value was 1 R per 10¹⁰ n/cm² from a criticality assembly (8). The TLD 700 reading was corrected for the effect of neutrons in Tab. 10.

The summary of the results from all gamma dose measurements are presented. in Tab. 11. A comparison with other detectors shows that the TLD 700 results agree sufficiently with the results of the other dosimeters. Nevertheless they are unsatisfactory due to the detailed correction in the different energy ranges and the remained discrepancy of the results. Further work 1s necessary to improve the corrections for the effect of intermediate neutrons, and/or the influence of other activation detectors, although no

aetivation foils from our own criticality dosimeter were exposed nearby the LiF dosimeters.

Using phosphate glasses only one correction for slow neutrons was neeessary, which can be done with high precision based on the activity measurements of 32_P of the same glass.

Acknowledgments

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References

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10^{10} PER ATOMS REACTIONS						
DETECTOR	PULSE 1	PULSE 2	PULSE ₃			
$197_{\rm Au}$ bare	$3,0.10^{-2}$	1.96 \cdot 10 ⁻²	$3.75 \cdot 10^{-2}$			
197_{Au} in In, Cd	$2,07 \cdot 10^{-2}$	$1.41 \cdot 10^{-2}$	$1.45 \cdot 10^{-2}$			
32 _s bare	$6 \cdot 10^{-5}$	$1.06 \cdot 10^{-5}$	$0.96 \cdot 10^{-5}$			
$31_{P(n,p)}31_{Si}$ +)	$3.22 \cdot 10^{-5}$	$0,455.10^{-5}$	$0.43 \cdot 10^{-5}$			
$31_{P(n,y)}32_{P}$ +)	$3.48 \cdot 10^{-7}$	$2,45.10^{-7}$	$3.6 \cdot 10^{-7}$			
76^{As} bare	$1.15 \cdot 10^{-3}$	$0,68.10^{-3}$	1.04.10 ⁻³			
76 As in Cd	$7.3.10^{-4}$	$4.35 \cdot 10^{-4}$	$2.65 \cdot 10^{-4}$			
237 $_{\mathrm{Np}}$ bare	$1.58 \cdot 10^{-3}$	$6.3 \cdot 10^{-4}$	$2.12 \cdot 10^{-4}$			
232 $_{\rm Th}$ bare	$6.3 \cdot 10^{-5}$	$1.9 \cdot 10^{-5}$	$1.2 \cdot 10^{-5}$			

Table 1: Reaction Rates for Personnel Dosimeter exposed in Free Air

+) Phosphate glass in boron sphere (Routine γ -Dosimeter)

Table 2: Fluence Estimates for Activation Deteetors

+) Au eovered by In and Cd

++) Arsenic inside Cd

+++) Phosphate glass dosimeter (Routine Y-dosimeter) in the spherieal capsule inside of a boron loaded plastic sphere

Table 3: Fluence Estimates for Threshold Detectors

Table 4: Dose Assessments for the Personnel Criticality Dosimeter and the Phosphate Glass Dosimeter

	IRRADIATION				NEUTRON	DOSE IN	$h \cdot erg \cdot g^{-1}$	
PULSE	SHIELDING	LOCATION	Au, Np, U, S	Au, Np, S	Np	Makrofol recoils	Au, S	Phosphate glass
	none	Free air Phantom A Phantom B	352 407	346 448	350 438	357 348 21	343 396 72	363 397 68
2	STEEL	Free air Phantom A Phantom B	132 177	136 161	14 _o 175	92 97 21	(145) 61 84 (185) 51(123)	55 (130) 59 (157) 47 (112)
$\overline{3}$	LUCITE	Free air Phantom A Phantom B	5 _o 58	53 59	47 5 _o	53 55 38	59 68 52	53 64 57

Table 5: Summary of Dose Estimates

 \bar{z}

Table 6: Dose Estimates for Personnel Dosimeter System exposed in Free Air

+) Assumed dose at area dosimeter location

Table 7: Dose Estimates for Man Phantom exposed facing Reactor

+) Dose measurement at front surface

		Dosimeter Reading (rd)		
PULSE	Phosphate Glass	D (boron)	D(Sn)	$D(Sn)$ ⁺⁾
				D (boron)
1	$FD-1$	7 _o	84	1.2
	$RPL-V$	47.5	47.	0.99
\overline{c}	$FD-1$	24	30	1.25
	RPL-V	15	15	1.0
$\overline{3}$	$FD-1$	56	114	2.04
	RPL-V	39	41.5	1.06

Table 8: γ -Dosimeter Reading of Phosphate Glasses in the Spherical Capsule and in a Tin Capsule for Free Air-Station

+) Phosphate glass in 1.2 mm tin capsule and in the routine dosimeter capsule inside of a boron plastic sphere

Table 9: Dosimeter Reading of LiF-Dosimeter

 $\label{eq:2.1} \frac{1}{\sqrt{2\pi}}\int_{\mathbb{R}^3}\frac{d\mu}{\sqrt{2\pi}}\left(\frac{d\mu}{\mu}\right)^2\frac{d\mu}{\sqrt{2\pi}}\int_{\mathbb{R}^3}\frac{d\mu}{\sqrt{2\pi}}\frac{d\mu}{\sqrt{2\pi}}\frac{d\mu}{\sqrt{2\pi}}\frac{d\mu}{\sqrt{2\pi}}\frac{d\mu}{\sqrt{2\pi}}\frac{d\mu}{\sqrt{2\pi}}\frac{d\mu}{\sqrt{2\pi}}\frac{d\mu}{\sqrt{2\pi}}\frac{d\mu}{\sqrt{2\pi}}\frac{d\mu}{\$

Table 11: γ -Dose Estimates for the LiF-Dosimeter and the Phosphate Glass Dosimeter in the spherical capsule

+) corrected for thermal and fast neutrons, see Table

⁺¹) Routine personnel dosimeter: Toshiba FD-1 glass inside of a boron plastic sphere

+++) the same dosimeter capsule with Schott RPL-V glass, which has a low sensitivity to thermal neutrons.