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MEASUREMENT OF GAMMA DOSE RATES ON PACKAGES LOADED WITH SPENT FUEL ASSEMBLIES

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ABSTRACT

After the loading of spent fuel assemblies in a cask, and prior to shipment, measurements are performed to determine the maximum dose rates around the package and the conveyance. These maximum dose rates shall comply with regulatory limits specified in the regulation. The measurements are also compared with predicted values calculated with the spent fuel characteristics prior to the loading. Consignees are also required to perform dose rate measurements on the packages and conveyances. The measurements should therefore be conservative, while being as accurate as possible, with a good repeatability. The choice of a device for the measurement of gamma dose rates must be done carefully, as several important factors may have a strong influence on the result of the measurement, such as the reference spectrum for the calibration, the size of the sensible volume of the detector, the integration time, etc. The spectrum of emission of spent fuel assemblies may vary according to the type of fuel (enriched U or MOX), the enrichment, the burn-up, and the cooling time. Furthermore the spent fuel assemblies usually generate gamma photons of high energy. The device should be able to measure accurately the photon emission on a wide energy range. This paper presents an ORANO / EDF survey on some devices designed for the measurements of gamma radiations on spent fuel packages, including an inter-comparison with measures on ORANO TN casks loaded with EDF fuel. It appears that some devices may strongly overestimate the gamma dose rates due to the presence of high energy secondary gamma photons.

INTRODUCTION

During transport, the dose rates on packages, overpacks and conveyances must comply with the regulatory requirements of IAEA regulation [1]. Thresholds are defined for the maximal dose rates on the external surface of the package, overpack, freight container or vehicle, at a distance of 1m from the package (when the transport index is limited), and at a distance of 2m from the vehicle or freight container.

In order to measure the radiation level, an appropriate instrument should be used. The radiation meter should be sensitive to the type of radiation produced by the package, and calibrated for it. The measurements should therefore be conservative, while being as accurate as possible, with a good repeatability.

The spent fuel assemblies usually generate gamma photons of high energy. The radiation meter should be able to measure accurately the gamma photon on the whole energy range of emission.

This paper presents an ORANO and EDF survey on some devices designed for the measurements of gamma radiations on spent fuel packages. An inter-comparison of measures was performed at BLAYAIS Nuclear Power Plant (NPP) around an ORANO TN12/2 cask loaded with EDF fuel.

Neutron radiation measurements are not discussed in this article.

TN12/2 SPENT FUEL PACKAGE

In France, spent fuel assemblies are transported from EDF's NPPs to ORANO's reprocessing plant at La Hague in ORANO's TN12/2, TN13/2 and TN112 casks (Figure 1). These packages may carry UO_2 and mixed oxide UO_2 -Pu O_2 (MOX) spent fuel assemblies.



Figure 1. ORANO TN12/2 cask for the transport of spent fuel assemblies

The survey consists in measuring the gamma dose rates around an ORANO TN12/2 package loaded with 4 MOX and 8 UO₂ spent fuel assemblies, to be shipped from BLAYAIS NPP to ORANO La Hague reprocessing plant.

ENERGY SPECTRUM IN AROUND THE TN12/2

A preliminary spectrometry was carried out at a distance of 17m from the package. This distance was suitable to not saturate the detector (about 1μ Sv/h maximum) and to limit the stacking phenomenon. The package was placed on a trailer, under a tarpaulin.

For this purpose a CANBERRA FALCON 5000 was used. This device is equipped with a Broad Energy Germanium detector (Figure 2). It was calibrated with a ⁶⁰Co source (1.17 MeV and 1.33 MeV), a ¹³⁷Cs source (0.66 MeV) and a ²³⁸Pu/¹³C source (5.11 MeV, 5.62 MeV and 6.13 MeV). The calibration curve was extrapolated up to 8.6 MeV.



Figure 2. Spectrometry measures on an ORANO TN12/2 at BLAYAIS NPP

The background of the environment was measured on BLAYAIS NPP. The highest natural gamma ray is given by the ²⁰⁸Tl at 2615 keV.



Figure 3. Spectrum of energy at 17 m from the package compared to background

The spectrum obtained at the distance of 17 m from the package, after 120 minutes, shows the presence of gamma photons of high energy, certainly up to 8 MeV. The background of the environment is negligible compared with the signal generated by the spent fuel assemblies (Figure 3).

These high energy photons are mainly produced by - (n, γ) reactions occurring in the body of the packaging. In these reactions, the compound nucleus deexcites by gamma emission to the ground state. The cross-section of this kind of reaction becomes high when thermal neutrons interact with low Z materials, as it can be found in the body of the package.

The theoretical contribution of the gamma energy spectrum to the overall gamma dose equivalent rate at the surface of the TN12/2 package was calculated by ORANO TN. An example of the result of such calculation, for a "representative content", on a radial middle point at package surface, is shown on Figure 4.

In this specific example, photons with an energy below 3 MeV contribute for 68% to the total gamma dose equivalent rate on the surface of the package. Photons with an energy above 3 MeV contribute for 32% to the total gamma dose equivalent rate on the surface of the package, with the following approximate repartition:

- 4% for photons in the range 3 to 4 MeV,
- 8% for photons in the range 4 to 6 MeV,
- 20% for photons in the range 6 to 8 MeV.

It shall be noted that the energy distribution varies greatly with the content and type of package. However the range remains approximately the same (up to about 10 MeV).



Figure 4. Contribution to the total gamma dose equivalent rate of each range of energy (contact package)

SURVEY ON 10 DEVICES FOR THE MEASUREMENT OF GAMMA DOSE EQUIVALENT RATES

Devices used for gamma dose rate measurements are chosen to compare estimated dose rates under the same conditions with different types of detector: Geiger Müller, proportional counter, plastic scintillator, silicon detector, ionization chamber. The energy responses being different, a difference in the estimated gamma dose rate is thus expected.

The survey was conducted using various devices. Measurements were performed on ORANO's TN12/2 at BLAYAIS NPP. The characteristics of the devices are shown on Table 1. The technology, the supplier, the name, the measured quantity and the scope of each device are specified. The devices are meant for radioprotection (RP), environment (ENV), personal dosimetry (DOS) and for calibration (STA) purposes.

Technology	Manufacturer/ Supplier	Device Name	Scope	Measuring Quantity
	AUTOMESS/ SAPHYMO	6150 AD6/H	RP	
Geiger Müller	AUTOMESS/ SAPHYMO	6150 AD5/H	RP	Ambient dose equivalent
detector	SAPHYMO	MiniTrace gamma	RP	rate $\dot{H} * (10)$
	GENITRON/ SAPHYMO	Gamma Tracer	Env	
proportional counter detector	THERMO FISHER/ APVL	FH 40 GL-10	RP	Ambient dose equivalent rate $\dot{H}^*(10)$
Plastic	APVL	AT1123	RP-Env	Ambient dose equivalent
scintillator	AUTOMESS/ SAPHYMO	AD b	RP-Env	rate
Silicon detector	SAPHYMO	SGI	RP-DOS	Personal dose equivalent <i>H</i> p(10)
Ionization chamber	NARDEUX / CANBERRA	Babyline 81	RP	Absorbed dose rate $\dot{D}t(3)$
	PTW	UNIDOS 10 liters	STA	Kerma in the air K _{air}

Table 1. Characteristics of the devices used for the survey

The campaign at BLA NPP aims to compare the measured values of each device with respect to the operational quantity $\dot{H}^*(10)$, according to the international standard IEC 60846-1.

It should be noted that the different geometries of the detectors hinder a rigorous comparison of the measurements performed at the contact of the package. As an example, the ADb probe embeds a 3 inch x 3 inch cylindrical plastic scintillator. The measured dose rate refers at a distance detector-surface of about 8 cm.

The UNIDOS dosimeter is used as a standard in calibration laboratories. The measured physical quantity is the Kerma in the air (Kinetic Energy Released in MAterial) in Gy/h. This device is not suitable for measurements at the contact of the surface, due to the size of the detector. The measured values are corrected according to the ambient pressure and temperature. The operational quantity $\dot{H}^*(10)$ is recalculated on the basis of the knowledge of the energy spectrum and the application of the conversion factors Kerma / $\dot{H}^*(10)$ from the ICRP 74.

The Babyline 81 is equipped with a 517 cm³ ionization chamber. The measured physical quantity is the absorbed dose or dose rate under 3mm of equivalent tissue $\dot{D}t(3)$ in Gy or in Gy/h. The measured values are corrected according to the ambient pressure and temperature. The response of the Babyline is consistent with the operational quantity $\dot{H}^*(10)$ over a wide range of the energy measurement domain.

The personal dosimeter SGI is equipped with a silicon detector. The dose rate is calculated by normalising the integrated dose by the integration time (several minutes). The measured physical quantity is the personal dose equivalent rate $\dot{H}p(10)$ in Sv/h. The quantity $\dot{H}p(10)$ is very close to the quantity $\dot{H}^*(10)$ for the photons of high energy (see Figure 6).

The TN12/2 package was positioned horizontally on a trailer. The measurements were performed on specific points illustrated in Figure 5:

- on the right middle radial points (270°) at the surface of the package, at a distance of 1m from the package, and at a distance of 2m from the trailer;
- on the left middle radial points (90°) at the surface of the package, at the surface of the trailer, and at a distance of 2m from the trailer;
- on the left (90°) rear and front trunnion bases at the surface of the package and at a distance of 1m from the package.



Figure 5. TN12/2 at BLAYAIS NPP – Measurement points

The results of the measures are shown on Table 2 and Table 3. The given values are averaged on at least 10 instantaneous measures.

	Radial point (270°)			Radial point (90°)			
	Contact	1 m	2 m	Contact	Contact	2 m	
_	Package	package	trailer	Package	trailer	trailer	
	414,6	171,5	77,1	420,6	305,5	79,9	
AD6/H	±2,4	±3,8	±1,2	±2,7	±6,7	$\pm 1,8$	
MiniTrace γ	391,8	169,3	75	404,5	310	83	
•	±1,6	$\pm 1,8$		±3,2	±4	±1	
Gamma Tracer	-	159,83	69,6	-	-	71,54	
		±1,14	±0,72			±0,59	
AD5/H	340,8	135,3	52,9	323,9	260	63	
	±10,7	$\pm 4,8$	±5,8	±13,3	± 8	±4	
FH 40 GL-10	328,5	133,9	55,3	337,4	246	59,9	
	±5,8	±3,2	±0,6	±11,2	±6	$\pm 0,8$	
AT1123	255	108	44	258	185	48	
	±2,55	±2,16	$\pm 0,88$	±2,58	$\pm 1,85$	±0,96	
AD b	Saturation	95	44,5	-	Saturation	45	
SGi	243	108	44,3	224	175	45	
Babyline 81	248,2	98,8 µGy/h	42,9	243,5 µGy/h	175,1 µGy/h	41,9	
	µGy/h		µGy/h			µGy/h	
10 Liter Ionization	-	88	38,3	-	-	38,94	
Chamber		µGy/h	µGy/h			µGy/h	

 Table 2.
 Measured dose rates at different points, in µSv/h (unless otherwise stated)

Table 3.	Measured dose	rates at different	points, in	μSv/h	(unless	otherwise s	tated)
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	Left front trunnion (90°)		Left rear trunnion (90°)		
	Contact Package	1 m package	Contact Package	1 m package	
AD6/H	182,4	61,6	112,5	48,9	
	$\pm 1,1$	±1,2	±4,3	±1,2	
MiniTrace γ	183,7	60,2	112,6	49	
·	$\pm 1,8$	$\pm 0,8$	±1,7		
Gamma Tracer	-	59,07	-	-	
		$\pm 0,58$			
AD5/H	132,4	47,4	86,6	25,7	
	±9,3	±3,3	$\pm 9,5$	$\pm 4,1$	
	153,7	51,9	98	43,6	
FH 40 GL-10	±6,3	±0,4	±2	±3,4	
AT1123	106	40	63	31	
	±2,12	$\pm 0,80$	±1,26	±0,31	
AD b	93,4	35	-	30,6	
SGi	120	37,5	70	29,3	
Babyline 81	109,1 µGy/h	37 µGy/h	79,2 µGy/h	29,2 µGy/h	
10 Liter Ionization Chamber	-	35 µGy/h	-	27,2 µGy/h	

DISCUSSION

Many factors can explain the deviations noted in the measurements of the dose rates on the package. One part of these factors is directly linked to human performance, for instance correct positioning of the devices. This will not be discussed hereunder, as it is assumed that special care was taken during such measurements. This may however remain a point of vigilance during routine measurements on-site.

The first source of error comes from the devices measuring different physical quantities. The values presented in Tables 2 and 3 express different radiological operational quantities, according to the used device for the measurement:

- Kerma in the air,
- \circ absorbed dose rate under 3mm of equivalent tissue $\dot{D}t(3)$,
- ambient dose equivalent rate $\dot{H}^*(10)$,



 \circ personal dose equivalent rate under 10mm $\dot{H}p(10)$.

In the range of 10keV to 1MeV, the error is significant depending on the radiological physical quantity used by the device to estimate the ambient dose equivalent rate $\dot{H}^{*}(10)$, as shown in Figure 6.

The estimation of the ambient dose equivalent rate $\dot{H}^*(10)$ starting from physical quantities such as the Kerma in the air (Kair), or the absorbed dose rate under 3mm of equivalent tissue $\dot{D}t(3)$, leads to an error from -40% to + 80% in the energy range from 10 keV to 1 MeV (Figure 7).

This energy range, according to theoretical calculations (see Figure 4) performed for a "representative content" on a radial middle point at a TN12/2 package surface, induces about 35% to 40% of the total gamma dose in contact (68% in the range under 3 MeV). This leads to a theoretical significant underestimation of the dose equivalent rate, and therefore confirms the use of $\dot{H}^*(10)$ radiation meters to evaluate the regulatory values of the ambient dose equivalent rate at different distances from the package.



Figure 7. Error following a measurement $\dot{D}t(3)$ (normalised to 1 for the ¹³⁷Cs) for the quantity $\dot{H}^*(10)$ in comparison with the true value $\dot{H}^*(10)$. $L_{H^*(10)} = L_{Dt(3)} \cdot (L_{Dt(3)} \times \Delta/100)$

The second source of error comes from the sensitivity of $\dot{H}^{*}(10)$ radiation meters as a function to gamma photons energy.

Radiation meters currently used for the measurements on the spent fuel packages are $\dot{H}^{*}(10)$ radiation meters, in accordance with IEC 60846 international standard. This standard limits the variation of the relative response for gamma energies up to 1.5 MeV between -29% and +67%. There is no given variation limits for photon energies above 1.5 MeV.

The inter-comparison of equivalent dose rates measured under the same experimental conditions with radiation meters of different technologies confirms that their energy responses differ significantly in the range of 6 to 7 MeV (Figure 8).

The result is the following classification of radiation meters according to their technology:

- The Geiger Muller detector gives the highest dose rate, it overestimates the highest energies (especially the AD6/H).
- The proportional counter detector gives a value lower than the Geiger Muller, the high energies are less overestimated (FH 40 GL-10).
- The dose rate calculated with the dosimeter equipped with a silicon detector is close to the expected value with a slight overestimation since the value $\dot{H}p(10)$ used by this device is greater than the $\dot{H}^*(10)$ value used by the radiation meters. The energy response is very little dependent on incidental gamma energy.
- Dose rates measured by plastic scintillator detectors are equivalent to those measured with silicon detector. The energy response is not much fluctuant for the ADb, but the dose rate is underestimated by the AT1123 for the highest energies.

 \circ Ionization chambers give values (Kerma in the air) close to the dose rates measured with the silicon detector, as there is a large presence of high energy photons for which the coefficients $\dot{H}p(10)/K_{air}$ are close to 1.1. The energy response of the ionization chambers is quasi constant.



Figure 8. Response in energy

CONCLUSION AND PERSPECTIVE

Radiation meters equipped with Geiger-Müller detectors are commonly used on French NPPs. They comply with IEC 60846 international standard requirements, and are suitable for the measurements of gamma ambient dose equivalent rates on most workplaces and environments in the facilities. It was shown that these detectors may strongly overestimate the ambient dose equivalent rate for the high energy gamma photons present next to a package loaded with spent fuel assemblies.

It was agreed that the radiation meter FH40 GL-10, with its proportional counter detector, presents an energy response within the limits defined by IEC 60846 international standard, together with a range of use that was suitable for measures of the gamma effective dose rates on the packages used for the transport of spent fuel assemblies. This radiation meter is now used on French NPPs and at ORANO's facilities for the shipment of spent fuel assemblies packages.

Nevertheless this device overestimates the ambient effective dose rate for the energies above 4.4 MeV.

Two technologies could optimise the measurement of gamma ambient effective dose rates:

- Plastic scintillator detectors, which have the disadvantage of their fragility and their large volume, not suitable for the measurements on the surface of the packages and vehicles.
- Silicon detectors, which have a quasi-constant response in low and high energy. This technology is robust and miniature, suitable for the measurements on contact of the surfaces. Currently no manufacturer offers this technology on radiation meters.

This conclusion does not call into question the quality of all other devices, especially those mentioned in this paper. In any case, it is important to choose a device suitable to the gamma energy range involved.