

Yearly criticality dosimetry test 2014

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1 Introduction

In places where fissile materials are being handled and processed, human error or failure of the safeguards system could lead to a criticality accident during which a critical mass or volume of fissile material is being reached. Although such accidents almost stopped occurring as a result of the high level of today's safety technology, the risk of a criticality accident can never be absolutely eliminated.

Criticality accidents require specialized neutron dosimetry techniques, which differ markedly from those used in routine radiological protection. This is mainly caused by the fact that one should be able to measure high doses in neutron fields with very high instantaneous dose rates. Dose values assumed to be of general concern for criticality dosimetry are in the range from 250 mGy up to 10 Gy. The dose rates may be up to 10^5 Gy/s.

Specific requirements of a dosimetry system used in criticality accidents situations are described in Chapter 3 of IAEA Technical report series No. 211 (IAEA, 1982):

- In the first place, the technique must allow a quick separation of exposed and non-exposed persons after the accident. In the second place, the technique must be able to separate the neutron and the gamma component of the dose.
- Another feature of the criticality dosimetry system is that doses must be reconstructed within an uncertainty of less than 50% within 48 hours and less than 25% four days later, and this should be done for a broad dose range spanning from 100 mGy up to 10 Gy.
- Since the sensitivity of neutron detectors usually strongly depends on the neutron energy, the system must be able to reconstruct the neutron spectrum or at least estimate the average neutron energy.

2 SCK•CEN criticality dosimeters

The criticality dosimetry service provided by the SCK•CEN to their customers is accredited according to ISO17025 by the Belgian Accreditation Organisation BELAC. The service is based on dosimeters with a set of four activation detectors (^{197}Au with and without Cd cover, ^{115}In and ^{32}S). In case of irradiation with neutrons, the following nuclear reactions take place:

- $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$
- $^{115}\text{In}(n,n')^{115\text{m}}\text{In}$
- $^{115}\text{In}(n,\gamma)^{116\text{m1}}\text{In}$
- $^{32}\text{S}(n,p)^{32}\text{P}$

^{198}Au undergoes β -decay to $^{198}\text{Hg}^*$ with a half-life time of 2.7 days. Subsequently, $^{198}\text{Hg}^*$ undergoes γ -decay (412 keV) to its ground state with a half-life time of 23 ps. The activity of ^{198}Au can be measured by means of gammaspectrometry with a germanium detector. $^{115\text{m}}\text{In}$ undergoes γ -decay (336 keV) to its ground state with a half-life time of 4.49 hours. Also the activity of $^{115\text{m}}\text{In}$ can be measured by means of gammaspectrometry with a germanium detector. $^{116\text{m1}}\text{In}$ undergoes γ -decay (127 keV) to its ground state with a half-life time of 54 min. This short lived radionuclide can be used for fast separation of irradiated and non-irradiated people. The activity of $^{116\text{m1}}\text{In}$ can be measured with a simple dose rate meter in close contact with the dosimeter badge. A neutron dose of 0.1 Gy gives 10 minutes after irradiation a dose rate of roughly 6 $\mu\text{Sv/h}$. ^{32}P undergoes β -decay (1.71 MeV) with a half-life time of 14.3 days. The activity of ^{32}P can be determined by means of liquid scintillation counting after chemical separation from ^{32}S .

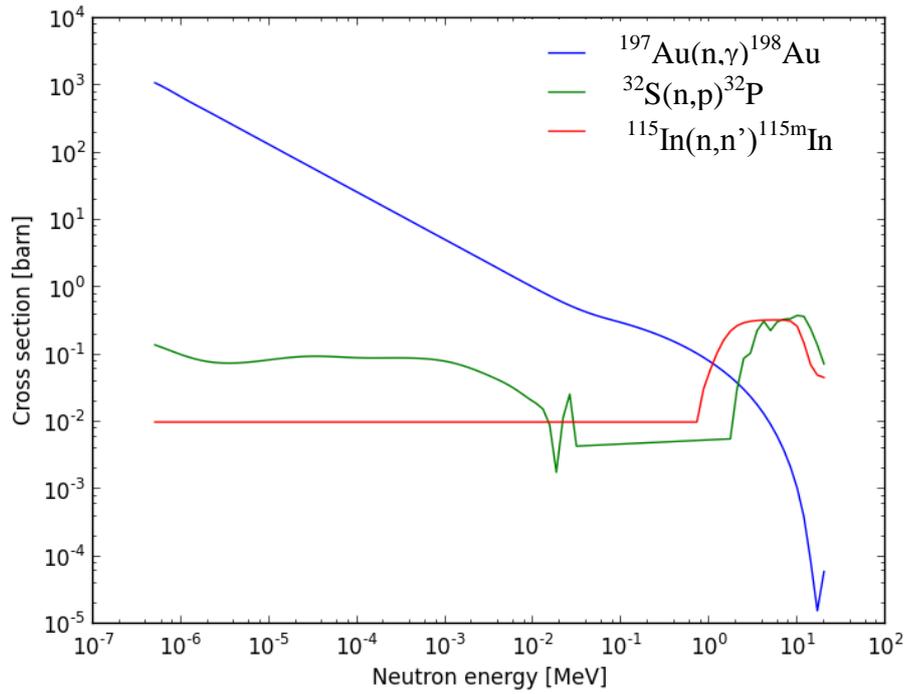


Figure 1: Neutron activation cross sections of the reactions used for reconstructing the neutron energy spectrum.

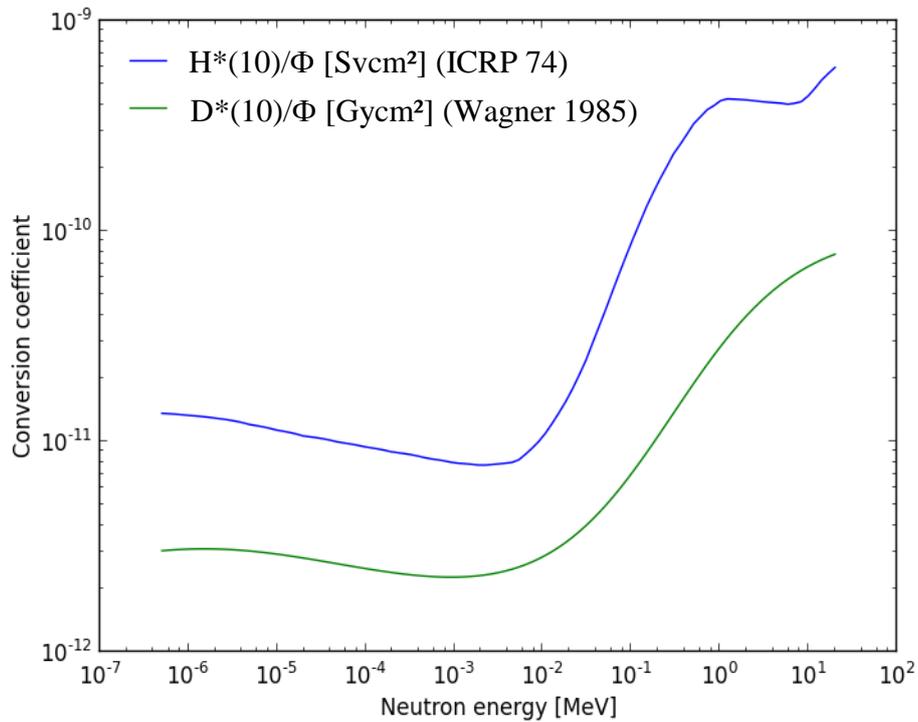


Figure 2: Energy dependence of the $H^*(10)/\Phi$ and $D^*(10)/\Phi$ conversion coefficients

The thermal neutron fluence below the Cd cut-off can be calculated from the difference between the measured activities of ^{198}Au in the gold foils without and with Cd cover. The measured activities of ^{198}Au in the gold foil without Cd cover, $^{115\text{m}}\text{In}$ and ^{32}P allow recovering the victim's dose in terms of $H^*(10)$ and $D^*(10)$. First the neutron fluence energy spectrum is reconstructed. This is possible due to the different energy dependence of the neutron activation reactions as shown in figure 1. The fluence energy spectrum is reconstructed by means of an iterative calculation starting with a realistic initial fluence energy spectrum as presented in Doroshenko et al. (1977). The doses in terms of $D^*(10)$ and $H^*(10)$ can then be calculated from the fluence energy spectrum by using the $D^*(10)/\Phi$ and $H^*(10)/\Phi$ conversion coefficients shown in figure 2.

Recently a new calculation tool with an easy-to-use graphical user interface was developed to automate the dose calculations. The user only has to plug in the measured activities with the corresponding uncertainties and select the appropriate predefined initial spectrum (^{235}U fission or combined spectrum) or provide another spectrum by means of a text file. The tool then reconstructs the energy spectrum and the dose in terms of $H^*(10)$ and $D^*(10)$ with the corresponding uncertainty range. The tool allows a much easier calculation of the doses and excludes calculation errors.

The advantage of the current criticality dosimetry system at SCK•CEN is that there is no need for maintenance of the dosimeters or periodic read-out. The dosimeters do not return to SCK•CEN's dosimetry service, unless an accident occurs.

3 Description of the experiments

Since our criticality dosimeters do not require any maintenance, a yearly test is incorporated in the quality assurance program. The main goal of this test is to avoid loss of knowledge in the different laboratories and to validate the dose reconstruction technique.

For the test conducted on August 28 2014, 2 dosimeters were irradiated to a known dose in the central cavity with uranium sphere in the BR1 reactor. After irradiation the sulphur pellets and gold and indium foils were sent for analysis to the responsible laboratories. Sulphur was analysed by the laboratory for liquid scintillation counting, while the gold and indium foils were analysed by the laboratory for gamma spectrometry. Both laboratories are part of the Expert Group for Low Level Radioactivity Measurements. These activity measurements are also part of the ISO17025 accreditation. The results were then used to calculate the neutron fluence energy spectrum and the doses in terms of $D^*(10)$ and $H^*(10)$ with the new calculation tool. The calculations were performed independently by three people of the Expert Group Radiation Protection Dosimetry and Calibration.

4 Results

Dosimeters 1 and 2 were irradiated in the BR1 reactor for periods of respectively 20 s and 45 s, resulting in received doses of respectively 0.18 Gy (2.4 Sv) and 0.40 Gy (5.4 Sv). To confirm the possibility of a triage of exposed from non-exposed people via the activation of $^{116\text{m}}\text{In}$, a measurement was done with a Radiagem in close proximity of the criticality dosimeter at different times after irradiation. The results are presented in Table 1. These results confirm the fact that a dose rate of roughly 6 $\mu\text{Sv/h}$ can be measured when the dosimeter is exposed to a neutron dose of 0.1 Gy.

Table 1: Results of the measurements with a Radiagem in close proximity of the criticality dosimeter at different times after irradiation.

Dosemeter 1			Dosemeter 2		
Time after irradiation [min]	dose rate [$\mu\text{Sv/h}$]	Dose rate per 0.1 Gy [$\mu\text{Sv/h}$]	Time after irradiation [min]	dose rate [$\mu\text{Sv/h}$]	Dose rate per 0.1 Gy [$\mu\text{Sv/h}$]
1	15	8.3	2	44	11
5	12	6.7	6	22	5.5
8	12	6.7	8	21	5.3
21	9	5.0	20	16	4

Table 2: Measured activities of the activation detectors at the moment of the irradiation in Bq/g

Dosemeter	Au	Au(Cd)	In	S
1	1050 \pm 140	330 \pm 50	135 \pm 24	1.44 \pm 0.15
2	1580 \pm 240	670 \pm 100	320 \pm 50	3.1 \pm 0.3

Table 3: Calculated neutron doses compared to the reference values

Initial spectrum	Detector 1			
	$H^*(10)_{\text{calc}}$ [Sv]	$H^*(10)_{\text{calc}}/H^*(10)_{\text{ref}}$	$D^*(10)_{\text{calc}}$ [Gy]	$D^*(10)_{\text{calc}}/D^*(10)_{\text{ref}}$
BR1	2.22 (1.85-2.59)	0.92	0.17 (0.14-0.20)	0.95
^{235}U	3.03 (2.58-3.47)	1.26	0.23 (0.20-0.27)	1.31
Combined	1.74 (1.44-2.04)	0.73	0.14 (0.12-0.17)	0.80
Reference	2.40		0.18	
Initial spectrum	Detector 2			
	$H^*(10)_{\text{calc}}$ [Sv]	$H^*(10)_{\text{calc}}/H^*(10)_{\text{ref}}$	$D^*(10)_{\text{calc}}$ [Gy]	$D^*(10)_{\text{calc}}/D^*(10)_{\text{ref}}$
BR1	5.17 (4.41-5.93)	0.96	0.39 (0.34-0.45)	0.99
^{235}U	6.88 (5.99-7.76)	1.27	0.54 (0.46-0.62)	1.35
Combined	4.09 (3.47-4.72)	0.76	0.33 (0.28-0.38)	0.83
Reference	5.40		0.40	

The activities and corresponding uncertainties from the laboratory for liquid scintillation counting and the laboratory for gamma spectrometry are presented in Table 2. Table 3 compares the calculated total neutron doses in terms of $H^*(10)$ and $D^*(10)$ with the reference values. The uncertainty ranges of the calculations are shown in parentheses. The calculations were performed for three different initial neutron fluence energy spectra. The BR1 spectrum is the simulated spectrum for the irradiation configuration in BR1 used for this test. The ^{235}U spectrum is the pure ^{235}U fission spectrum. The combined spectrum is a combination of a 1/E spectrum and a ^{235}U fission spectrum. As expected the calculation performs best with the simulated BR1 spectrum. The calculated doses differ less than 10% from the reference doses which fall nicely within the uncertainty range of the calculations. With the ^{235}U spectrum the doses are slightly overestimated, while with the combined spectrum the doses are slightly underestimated. However, the deviations from the reference doses are always below 35%, which is well within the requirements of IAEA.

Figure 3 shows the three initial fluence energy spectra used for the iterative dose calculation (full lines) and the corresponding guessed spectra obtained from the calculation (dashed lines) for the two dosimeters. As expected, the plots show that the guessed spectra are very similar for the three initial spectra and closest to the simulated BR1 spectrum. This explains why the simulated BR1 spectrum gives the best agreement between the calculated and reference doses.

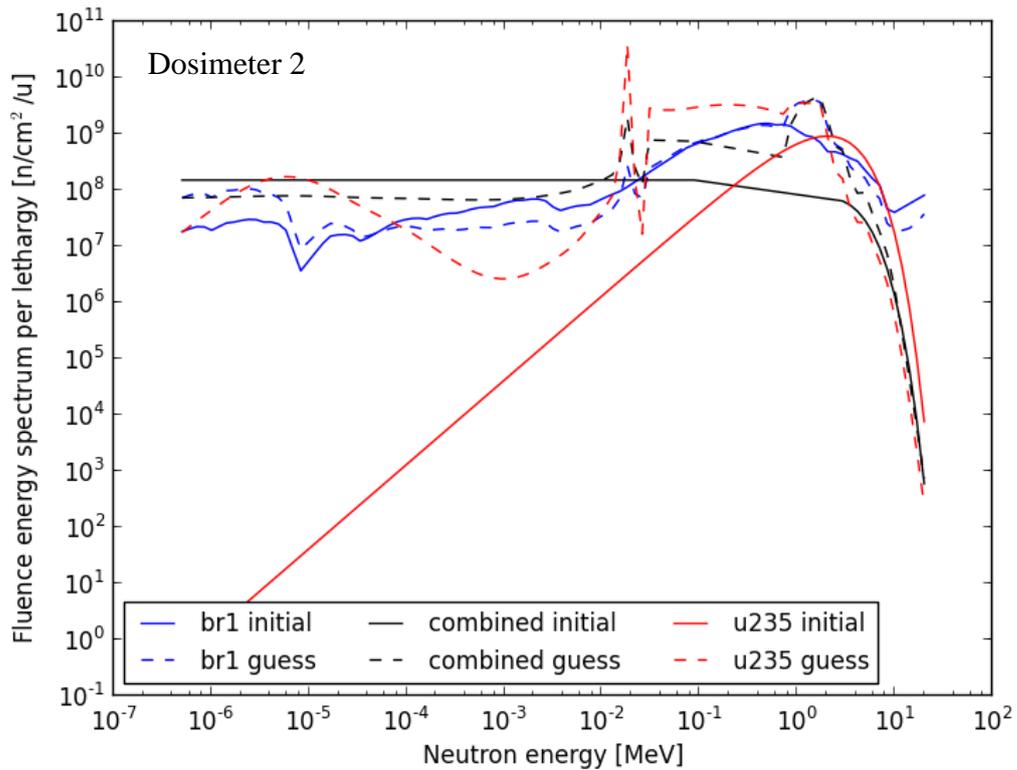
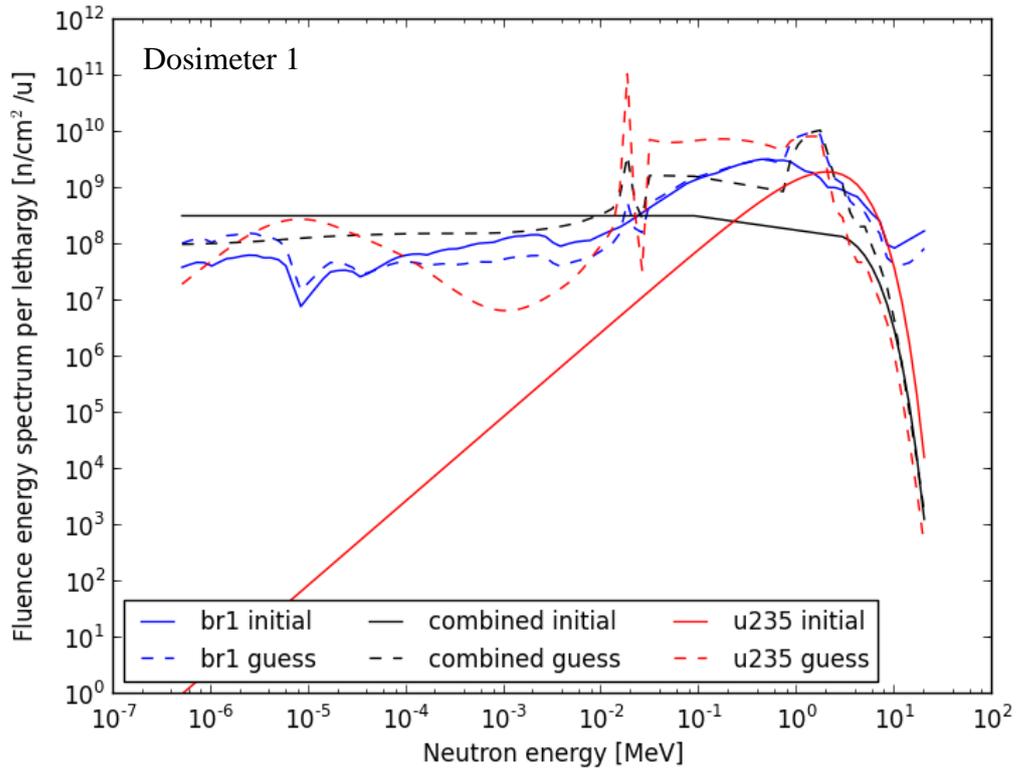


Figure 3: The three initial fluence energy spectra used for the iterative dose calculation (full lines) and the corresponding guessed spectra obtained from the calculation (dashed lines) for the two dosimeters

4.1 Conclusions

The yearly criticality test in the BR1 reactor confirmed the quality of the criticality dosimetry system. The test was conducted without irregularities. The calculated doses for both doseimeters approached the reference values within an uncertainty window of 35% with the general ^{235}U fission or the combined spectrum, while use of the more realistic simulated BR1 spectrum gave less than 10% deviation. This is well within IAEA requirements.