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CENTRE D'ETUDE DE L'ENERGIE NUCLEAIRE

YEARLY CRITICALITY DOSIMETRY TEST 2016

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Radiation Protection Dosimetry and Calibration

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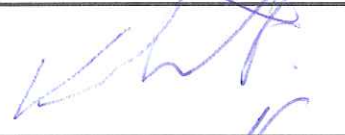

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1 Introduction

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 105 Gy/s.

Specific requirements of a dosimetry system used in criticality accidents situations are described in Chapter 3 of the IAEA manual on criticality dosimetry [1]:

- 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 ISO 17025 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 .

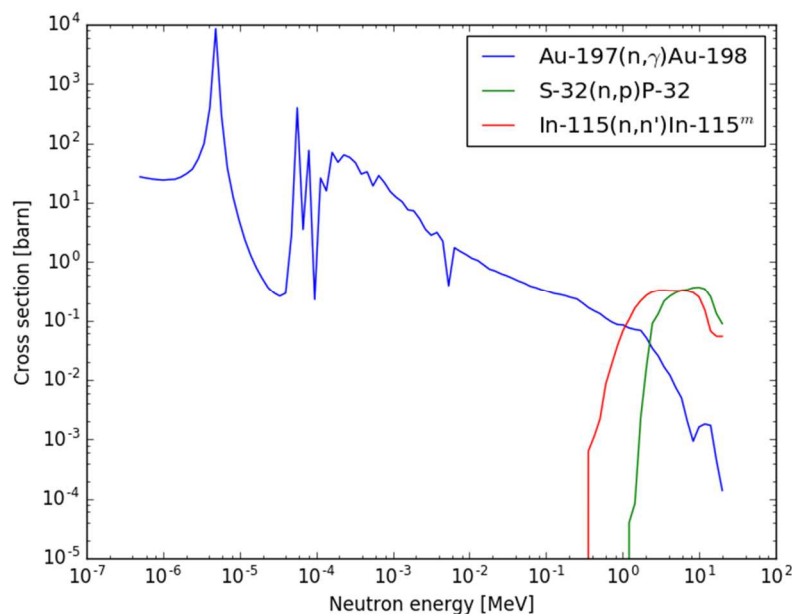


Figure 1: Neutron activation cross sections of the relevant reactions

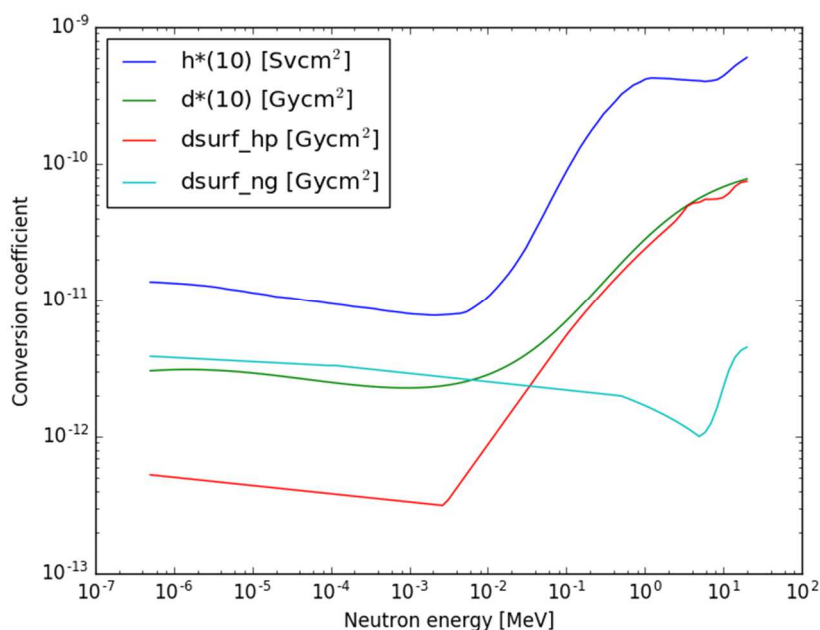


Figure 2: Energy dependence of the different dose conversion coefficients

The thermal neutron fluence below the Cd cut-off can be calculated from the difference between the measured activities of ¹⁹⁸Au in the gold foils without and with Cd cover. The measured activities of ¹⁹⁸Au in the gold foil without Cd cover, ^{115m}In and ³²P allow recovering the victim's dose in terms of the surface absorbed dose which is the quantity of interest for criticality accidents according to IAEA manual on criticality dosimetry [1]. 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 a publication by Doroshenko et al. [2]. The doses in terms of the surface absorbed dose can then be calculated from the fluence energy spectrum by using the dose conversion coefficients from the IAEA manual on criticality dosimetry [1]. There are separate conversion coefficients for the surface absorbed dose due to heavy charged particles and due to neutron induced gammas, respectively $d_{surf, hp}$ and $d_{surf, ng}$. This separation is necessary because the gamma component is already taken into account by the conventional gamma whole body dosimeter that should be worn as well. Hence, we are only interested in the heavy charged particle component. The energy dependence of these dose conversion coefficients is plotted in figure 2 together with that of $h^*(10)$ and $d^*(10)$ for comparison.

A 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 (²³⁵U 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 the surface absorbed dose 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 April 13 2016, 2 dosemeters were irradiated to a known dose in the central cavity of the BR1 reactor. The cavity was exceptionally not loaded with the 6 cm uranium shell such that the neutron energy spectrum was now purely thermal. 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 ISO 17025 accreditation. The results were then used to calculate the doses in terms of the surface absorbed dose.

4 Results

Dosemeters 1 and 2 were irradiated in the BR1 reactor for periods of respectively 9 min 32 s and 14 min 19 s. This results in surface absorbed doses due to heavy charged particles of respectively 268 mGy and 402 mGy or ambient dose equivalents of respectively 5 Sv and 7.5 Sv.

To confirm the possibility of a triage of exposed from non-exposed people via the activation of $^{116m1}\text{In}$, a measurement was done with a ambient gamma monitor in close proximity of the criticality dosimeter. The measured gamma dose rate ranged from 200 to 700 $\mu\text{Sv/h}$ per 0.1 Gy of surface absorbed dose. This is much higher than the typical value of 6 $\mu\text{Sv/h}$ per 0.1 Gy for the previous yearly tests. This is because the neutron energy spectrum used during this test was purely thermal and the $^{115}\text{In}(n,\gamma)^{116m1}\text{In}$ reaction responsible for most of the measured gamma emission has a much higher cross section for thermal neutrons.

Table 1: Measured activities of the activation detectors at the moment of the irradiation in Bq/g

Dosemeter	Au	Au(Cd)	In	S
1	295 000 \pm 40 000	270 \pm 50	< 80	< 0.27
2	450 000 \pm 65 000	410 \pm 60	< 310	< 0.4

Table 2: Calculated versus reference surface absorbed dose values

Dosemeter	Irradiation time	Reference dose [mGy]	Calculated dose [mGy]	Deviation [%]
1	9 min 32 s	268	243 (210 – 276)	- 9%
2	14 min 19 s	402	370 (317 – 424)	- 8%

The activities and corresponding uncertainties from the laboratory for liquid scintillation counting and the laboratory for gamma spectrometry are presented in table 1. As expected for a purely thermal neutron spectrum, the activity of the gold foil is very high and the activities of the In and S are not measurable.

The activities of the gold foil and the cadmium covered gold foil were then used to calculate the thermal neutron dose. The epithermal and fast neutron dose could be neglected because it was a purely thermal neutron spectrum. The calculated surface absorbed doses are compared with the reference surface absorbed doses in table 2. The 95% confidence interval are given between parentheses. The calculated values differ less than 10% from the reference values and the reference values fall nicely within the 95% confidence interval of the calculation. The half width of 95% confidence intervals are less than 15%. This is all well within IAEA requirements.

5 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 dosimeters deviate less than 10% from the reference values and the half widths of the 95% confidence intervals are less than 15%. This is well within IAEA requirements, requesting a dose estimate with less than 25% uncertainty.

6 References

- [1] IAEA Technical report series No. 211 (IAEA, 1982)
- [2] J.J. Doroshenko et al., Nuclear Technology, Vol.33 (May 1977)