Radiation Protection Dosimetry (2007), Vol. 124, No. 4, pp. 319–326
Advance Access publication 10 May 2007
doi:10.1093/rpd/ncm216

NEUTRON MEASUREMENTS AROUND STORAGE CASKS CONTAINING SPENT FUEL AND VITRIFIED HIGH-LEVEL RADIOACTIVE WASTE AT ZWILAG

T. Buchillier*, A. Aroua, and F. O. Bochud
University Institute of Applied Radiation Physics, Grand-Pré 1, 1007 Lausanne, Switzerland

Received December 11 2006, amended February 20 2007, accepted March 5 2007

Spectrometric and dosimetric measurements were made around a cask containing spent fuel and a cask containing high-level radioactive waste at the Swiss intermediate waste and spent fuel storage facility. A Bonner sphere spectrometer, an LB 6411 neutron monitor and an Automess Szintomat 6134A were used to characterise the n–γ fields at several locations around the two casks. The results of these measurements show that the neutron fluence spectra around the cask containing radioactive waste are harder and higher in intensity than those measured in the vicinity of the spent fuel cask. The ambient dose equivalents measured with the LB 6411 neutron monitor are in good agreement with those obtained using the Bonner spheres, except for locations with soft neutron spectra where the monitor overestimates the neutron ambient dose equivalent by almost 50%.

INTRODUCTION

In the absence of a high-level nuclear waste repository, which is still to be built in Switzerland when the investigation for an appropriate site will be finished, an intermediate waste and spent fuel storage facility (*Zwischenlager*—ZWILAG) has been constructed. It opened officially in the year 2000 and was granted a licence for operation in 2001. The ZWILAG facility is located in Würenlingen in the northern part of Switzerland, a region that hosts three of the four Swiss nuclear power plants. ZWILAG serves as a storage facility for spent fuel as well as all forms of radioactive waste in addition to the treatment of low-level waste from nuclear power plants, medicine, industry and research. It is owned by the four Swiss nuclear power plants: Beznau, Mühleberg, Gösgen and Leibstadt, which totalise a 3.25 GWe production(1).

The high-activity storage room in ZWILAG is able to accommodate 200 casks coming from the Swiss nuclear power plants, containing either spent fuel assemblies or vitrified waste. The Swiss federal nuclear safety inspectorate mandated the Lausanne University Institute of Applied Radiation Physics (IRA) who had already performed in the past several spectrometric and dosimetric measurements around different types of spent fuel storage casks in Switzerland(2) and Sweden(3), to characterise the neutron ambient dose equivalent rates in this storage room. A first set of measurements was undertaken in ZWILAG around the first cask stored in this room (TN97L type), filled with spent fuel coming from the Leibstadt nuclear power plant (KKL), with an estimated global activity of 242 PBq. A second set of measurements was undertaken in the vicinity of the first vitrified high-level radioactive waste cask (CASTOR HAW 20/28 CG type), filled with spent fuel from the Gösgen nuclear power plant (KKG) reprocessed in La Hague. The aim of the measurements described here is to characterise the neutron spectrum around the casks and to estimate the neutron ambient dose equivalent rate using a Bonner sphere neutron spectrometer and an integral instrument.

The results are compared with published studies concerning similar measurements around CASTOR IIa casks(4), CASTOR 440/84(5), CASTOR HAW 20/28 CG casks(6) or TN17/MK2(7).

MATERIALS AND METHODS

The measuring systems

The Bonner sphere neutron spectrometer

The Bonner sphere neutron spectrometer used in this work consists of a miniaturised proportional counter filled with ^{3}He, sensitive to thermal neutrons, and a set of 11 polyethylene spheres of diameters ranging from 2 to 15 inches (5.08–38.1 cm). Measurements of the count rates are successively taken with the bare counter, with the bare counter shielded by a cadmium sheath, and with the polyethylene spheres. The full description of the spectrometer, the establishment of its response matrix and the test of its global performance in reference neutron fields are already published(8).

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*Corresponding author: thierry.buchillier@chuv.ch

The neutron fluence spectra were unfolded from the experimental data using the GRAVEL unfolding code based on an improved SAND-II algorithm(9). The program provides the distribution of the neutron fluence with respect to the energy ϕ_E (spectrum), the total neutron fluence, and the ambient dose equivalent $H_n^*(10)$ using the fluence-to-dose equivalent conversion factors h_ϕ^* from ICRP74(10). In addition to the statistical uncertainty on the count rates, the unfolding procedure introduces additional uncertainty in the spectrum shape. However, the integral quantities (total fluence, ambient dose equivalent) are less affected. On the basis of computer simulations, calibrations and intercomparisons, the standard uncertainty on the dose is estimated at $\pm 10-15\%$.

The neutron and gamma ambient dosemeters

Neutron ambient dose equivalent rates were also measured with an LB 6411 (EG&G Berthold) neutron monitor associated with the LB 123 electronic. The LB 6411 is made of an ^{3}H/methane proportional counter located inside a 250 mm spherical low-density polyethylene moderator. The calibration is made by the manufacturer using a non-moderated ^{252}Cf neutron source. The energy range of this detector is 0.025 eV to 20 MeV. Gamma ambient dose equivalent rates were measured with an Automess Szintomat 6134A (scintillation detector for the energy range 25 keV to 1.3 MeV and dose rate 0.1 μSv h^{-1} to 0.1 Sv h^{-1}, calibrated in the ^{137}Cs photon radiation).

The measurement locations

The first set of measurements took place in the storage room for high-activity waste, close to the first and only cask present in this room, filled with spent fuel (Figure 1). Two spectrometric measurements were performed 1.0 m away from the cask at different heights and one measurement was performed at the centre top, 0.47 m above the cask (Figure 2). Finally, one spectrometric measurement was performed at a distance of 10 m away from the cask at a location where the ZWILAG radiation protection officers routinely perform integral measurements. The second set of measurements was performed around the first vitrified high-level radioactive waste cask from the Gösgen nuclear power plant (Figure 1). Two spectrometric measurements were performed 1.0 m away from the container at different heights and one measurement was performed at the centre top, 0.47 m above the container (Figure 2). An additional spectrometric measurement was performed at a distance of 10 m away from the container. Between the first and second sets of measurements, five casks filled with spent fuel and two filled with vitrified waste have been entered into the storage room. The second cask with vitrified waste from the Beznau nuclear power plant was located at a distance of 4 m only from the first one. Therefore, all measurements were taken on the opposite side with respect to the second cask in order to minimise its contribution. Table 1 shows the precise locations of the spectrometric measurement points. The casks consist of a thick-walled cylindrical body of 6.5-m height and 2.2-m diameter made of ductile cast iron. The wall thickness is approximately 0.4 m and includes polyethylene rods as a neutron moderator. A double-lid system and a shock absorber are bolted on top of the cask.

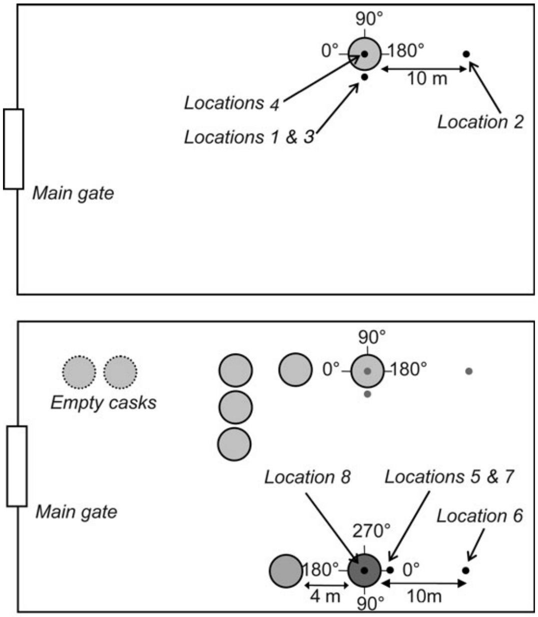

Figure 1. Schemes (not at scale) of the measurement locations around the measured casks. The first set of measurements (upper panel) was performed on a spent fuel cask. The second set of measurements (lower panel) was performed later around a cask containing a vitrified high-level radioactive waste.

RESULTS AND DISCUSSION

The Bonner sphere count rates

The readings of the Bonner spheres corrected for dead time are presented in Figure 3 as a function of the sphere diameter for the eight measurement locations. The statistical standard uncertainty shown in the figure is evaluated from the readings, assuming a Poisson distribution. The monotonic variation of the count rate versus sphere diameter provides an empirical check that no gross errors due to the

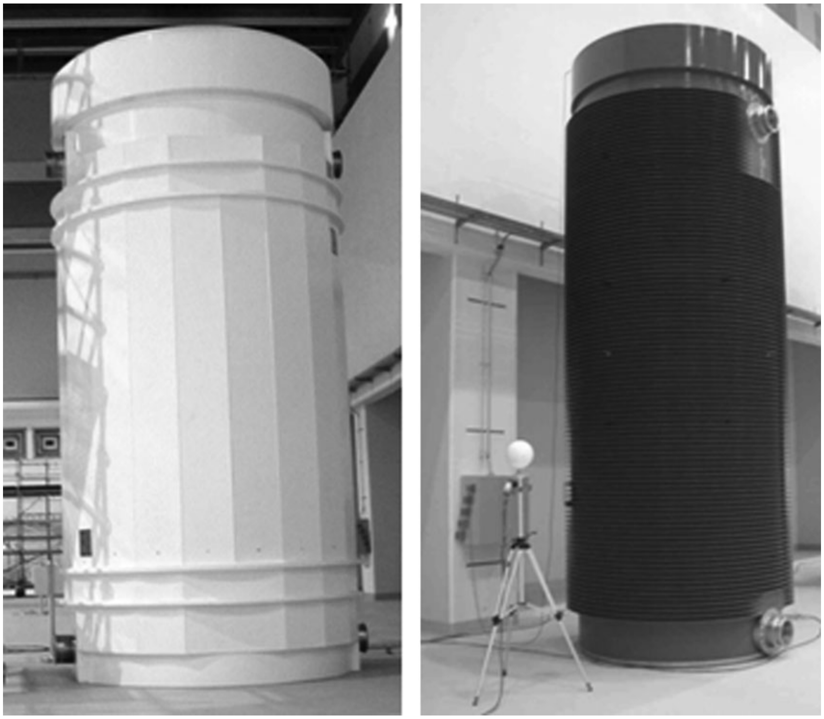

Figure 2. The casks investigated in this study, TN97L type containing spent fuel (left) and CASTOR HAW 20/28 CG type with vitrified high-level radioactive waste (right).

malfunction of the equipment occurred during the measurements(11).

In the absence of measurements with the 9-inch sphere for all the locations, the 9- to 3-inch ratio—a good measure of the neutron spectrum hardness(12)—could not be established. Instead, the 8- to 2-inch ratio was used. This ratio ranges from 0.53 for location 2 to 1.59 for location 7, as shown in Figure 3.

The neutron spectra

The dead time corrected count rates were unfolded with the response matrix of the Bonner spheres in order to obtain the neutron spectra. The SAND iterative procedure requires the specification of an initial guess spectrum. For all locations several guess spectra were considered. After the analysis of the various outputs, a starting spectrum of the following shape was adopted: a thermal distribution approximated by a constant fluence per unit lethargy from 0.01 to 0.1 eV, a constant fluence per unit lethargy until 1.97 MeV and then a ^{235}U fission tail. The resulting neutron fluence spectra are plotted in Figure 4 in the lethargy representation $E \cdot \dot{\phi}_{\mathrm{E}}$. They are essentially degraded fission spectra with a broad maximum around 100 keV.

From the spectra of Figure 4, the neutron fluence rates $\dot{\phi}_{\mathrm{n}}$ have been calculated and are displayed in Table 2. It can be seen that the fluence rate measured in the vicinity of the cask filled with vitrified high-level radioactive waste is about five times higher than that measured in the vicinity of the cask filled with spent fuel. Figure 4 also indicates that the neutrons fluence spectra associated with vitrified waste are harder than those associated with spent fuel, except for the top position (locations 4 and 8) where the spectrum shapes are quite similar, and the 8- to 2-inch ratios are close to each other (1.42 and 1.35, respectively). In Ref. (6) in contrast the authors observed a somewhat softer spectrum on top of a CASTOR V/19 cask than on the side wall. Also in this reference, the spectrum on top of a CASTOR V/19 with spent fuel was softer than on top of a CASTOR HAW 20/28 with vitrified waste. The difference was explained by the characteristic burn-up distribution of the spent fuel resulting in a low emission in the top and bottom regions. This distinction and the self-absorption by the fuel assembly top fittings resulted in a larger effective shielding and thus in a softer spectrum. In the present work, no information concerning the shielding of the spent fuel cask TN97L could be obtained. The above

Table 1. Locations of the measurements.

Location	Cask	Angle (°)	Distance from external surface (m)	Height (m)
1	Spent fuel	270	1	1.95
2	Spent fuel	180	10	0.86
3	Spent fuel	270	1	3.92
4	Spent fuel	On top	Centre	0.47 (above top surface)
5	Vitrified waste	0	1	1.95
6	Vitrified waste	0	10	1.95
7	Vitrified waste	0	1	2.89
8	Vitrified waste	On top	Centre	0.47 (above top surface)

explanation suggests that the construction of this type of cask is different from the CASTOR one.

Neutron fluence spectra at a distance of 1 m from the surface (locations 1 and 3, and locations 5 and 7) present similar shapes and 8- to 2-inch ratios (0.69–0.69 and 1.54–1.59). The lowest positions (1.95-m height) have spectra with a slightly higher contribution of intermediate neutrons that are probably scattered from the ground. The spectra at locations 4 and 8 (on top of the casks) are significantly harder. This is due to the fact that the lateral neutron shielding is probably thicker and thus the outcoming neutrons are more moderated laterally. On the other hand, the spectra at positions 2 and 6 (10 m away from the cask) are softer than those at positions 1/3 and 5/7. This is due to the contribution to this spectrum of neutrons scattered by the ground and the walls of the storage room.

The dosimetric results

The integral results obtained by the Bonner spheres and using the corresponding conversion factors are presented in Table 2. These are the average total neutron fluence rate $\dot{\phi}_n$ and the neutron ambient dose equivalent rate $\dot{H}_n^*$ (10) based on ICRP Recommendation 60(13). The gamma ambient dose equivalent rate $\dot{H}_\gamma^*$ (10) measured by the Automess Szintomat 6134A and the neutron ambient dose equivalent rate $\dot{H}_n^*$ (10) measured by the LB 6411 neutron monitor are also presented. The neutron ambient dose equivalent divided by the neutron fluence is given as a measure of spectral hardness at the different locations.

For each cask, the highest neutron ambient dose equivalent rates are registered at the lateral locations at 1 m distance from the surface of the cask (1/3 and 5/7). When comparing the neutron ambient dose equivalent rates at similar locations around the two casks, one observes that the cask filled with vitrified high-level radioactive waste registers a dose, $\dot{H}_n^*$ (10), 8.8 times higher than the cask filled with spent fuel.

Table 2 shows that the ratio between the gamma and neutron ambient dose equivalent rates can greatly vary. For instance, around the sides of the spent fuel cask, the photon dose component is higher with a neutron-to-photon ratio between 0.19 and 0.89. The greatest difference between the two components is found at the top of the container (location 4) where the neutron dose rate is about 25 times more important than the gamma. This is understandable by the fact that the top part of the container (mainly steel) is much thicker than the lateral parts and therefore specifically shields the photons. Conversely, the dose around the cask filled with vitrified high-level radioactive waste contains a dominant neutron component with a neutron-to-photon ratio ranging between 2.1 and 4.8 (2.1 on top). This ratio is very dependant on the type of cask, waste, history and measurement location and it is therefore difficult to make a comparison with the figures reported in the literature for other conditions. It can just be observed that the present values are in the same order of magnitude as those given in Ref. (5) on the side wall at 1 m height between groups of four CASTOR 440/84 casks with spent fuel and differ quite a lot for the top position (ratio of 6 and 23, respectively).

Figure 5 presents the ratio of the $\dot{H}_n^*$ (10) measurements performed by the LB 6411 neutron monitor and the Bonner sphere system with the 8- to 2-inch ratio. The uncertainties reported are obtained by the quadratic summation of the LB 6411 and Bonner sphere uncertainties. According to the informations given by the manufacturer and the periodic recalibration of the instrument by an authorised laboratory, the LB 6411 standard uncertainty is evaluated at ±6%. This is the uncertainty for the measurement in a radiation field of known energy distribution (^{252}Cf) and does not include the uncertainty due the energy dependence. If the latter is included, a total uncertainty of approx 30% or more is obtained. For the BS system, a standard uncertainty of ±15% is considered, which is the total uncertainty of the determination of $\dot{H}_n^*$ (10) in an

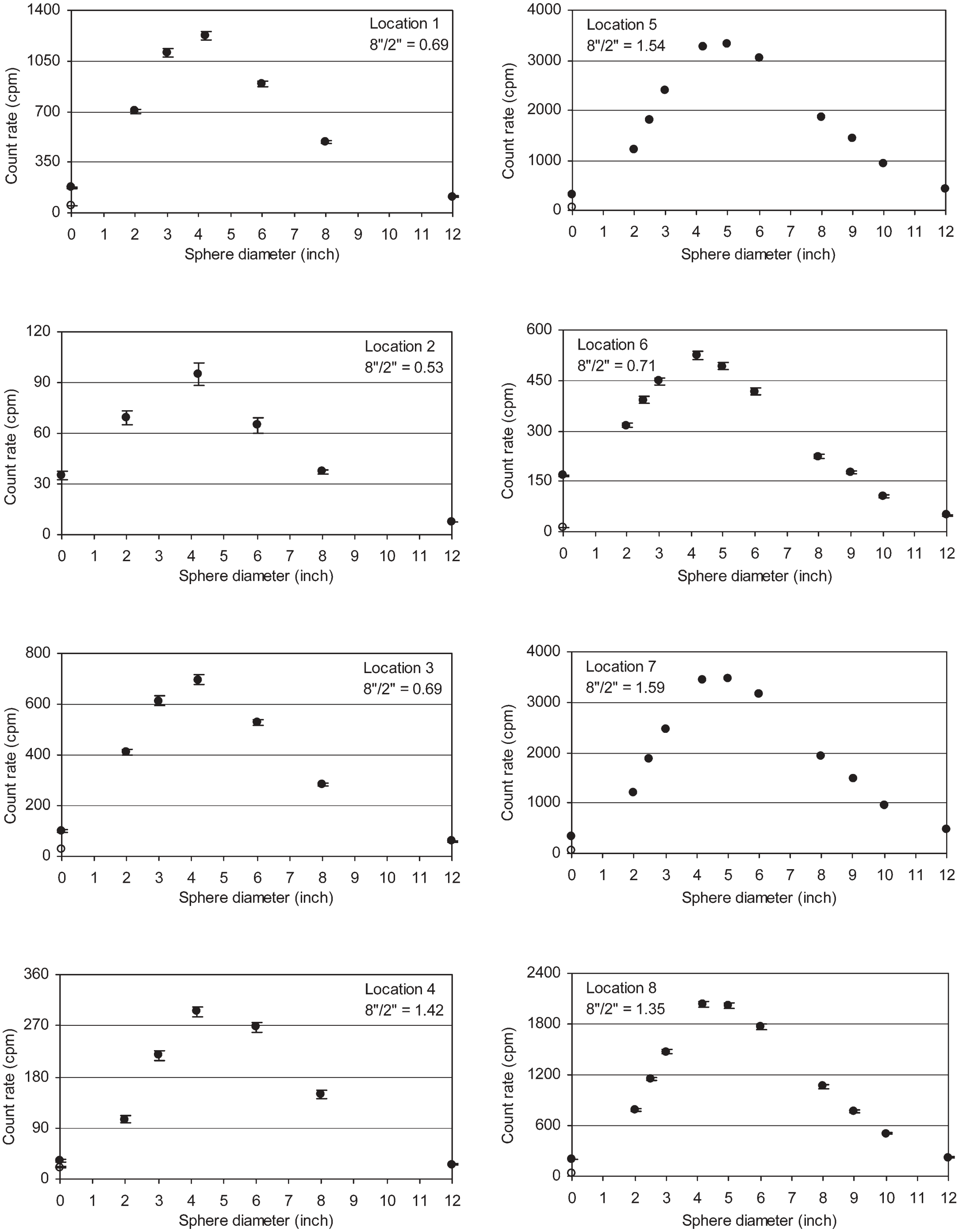

Figure 3. Count rates at the eight measurement locations at ZWILAG in the vicinity of a cask filled with spent fuel (left column) and a cask containing vitrified high-level radioactive waste (right column). The measurement with the cadmium-shielded counter is indicated by an empty circle.

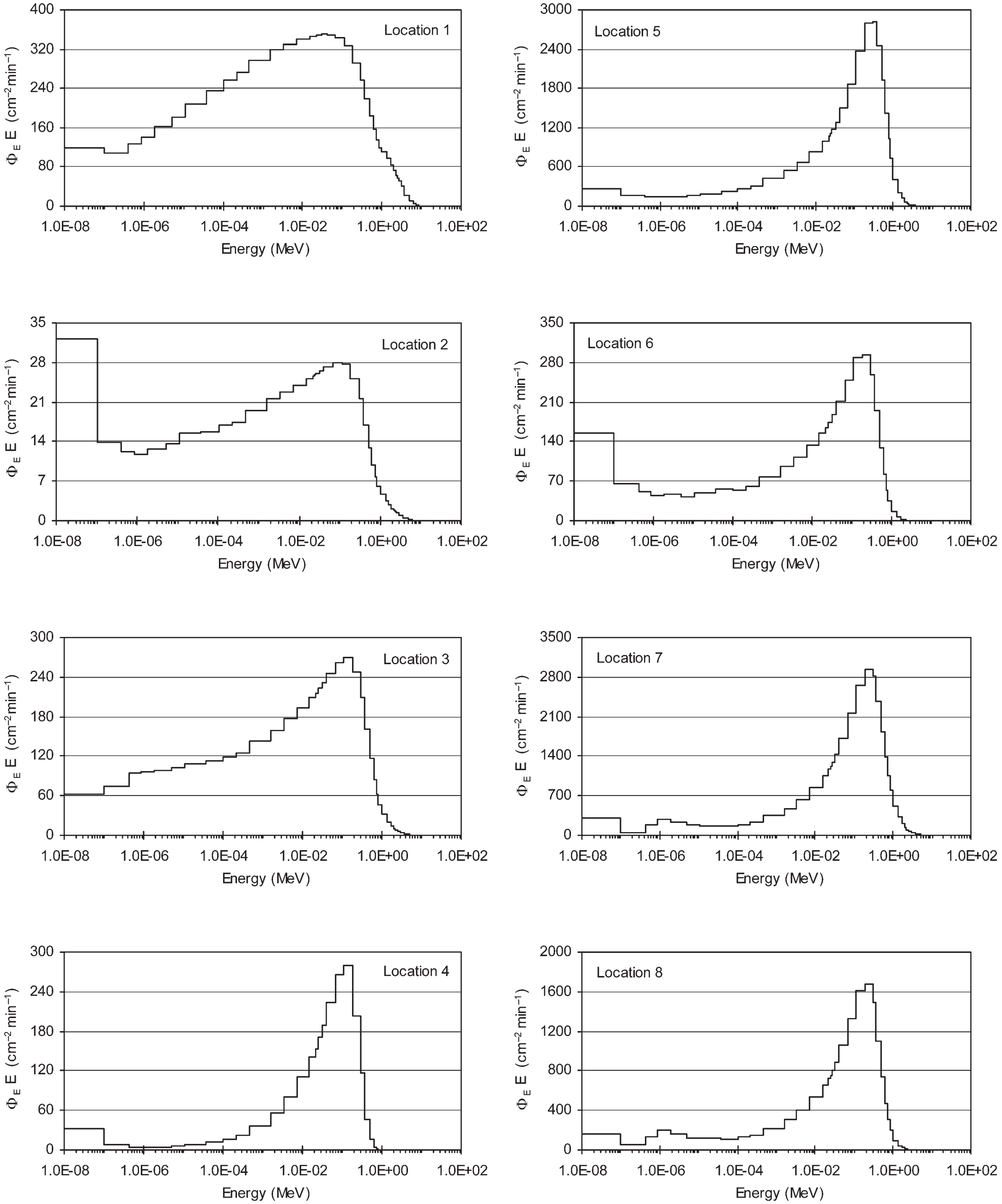

Figure 4. Neutron fluence spectra measured at the eight measurement locations at ZWILAG in the vicinity of a cask filled with spent fuel (left column) and a cask containing vitrified high-level radioactive waste (right column).

unknown radiation field. The use of the 6% uncertainty here is justified by the fact that the energy dependence is studied with the 8- to 2-inch ratio. The LB 6411 dose is higher than the Bonner spheres dose for soft spectra and lower for harder spectra. The LB 6411 neutron monitor overestimates $\dot{H}^*_n(10)$ by up to 40% for a spectrum with an 8- to 2-inch ratio of 0.5, and underestimates it by up to 20% for

Table 2. Detailed integral results at the various locations and standard uncertainties.

Location	$\dot{H}^*_\gamma$ (10) c(μSv h^{-1}) Szintomat ±10%	$\dot{\phi}_n$ (cm^{-2} min^{-1}) BS ±15%	$\dot{H}^*_n$ (10) (μSv h^{-1}) BS ±15%	$\dot{H}^*_n$ (10) (μSv h^{-1}) LB 6411 ±6%	Ratio LB 6411/BS ±16%	Ratio $\dot{H}^*_n$ (10)/ϕ_n (pSv cm^2)
1	15	4120	13.4	15.4	1.2	54
2	1.9	306	0.905	1.3	1.4	49
3	40	2390	7.78	8.8	1.1	54
4	0.17	1008	4.20	4.03	1.0	69
5	18	11640	81.8	74	0.9	117
6	3.5	1708	7.55	7.7	1.0	74
7	18	12020	86.5	76	0.9	120
8	19	6960	40.8	33	0.8	98

In the last column, the neutron ambient dose equivalent divided by the neutron fluence is given as a measure of the spectral hardness.

a spectrum with an 8- to 2-inch ratio of about 1.5. The overestimation for soft spectra is explained by the shape of the energy response curve of the LB 6411 given in the manual(14); in fact the instrument is particularly oversensitive (up to about a factor 10) for low-energy neutrons. From a radiation protection point of view, the performances of this instrument are quite satisfying because in the real situations where it is mostly used in the storage facility the indication is either conservative (around spent fuel cask or at some distance from the cask where scattered neutrons are present) or not too much underestimated (in the somewhat harder spectrum from vitrified high-level radioactive waste). This is also the conclusion of the EVIDOS study(7) on the basis of the spectra measured around a TN17/MK2 cask, which in fact show a larger contribution of intermediate and thermal neutrons than the spectra of the present work.

Figure 6 shows the readings of the neutron and photon area monitors, respectively, as a function of the distance to the surface of the cask. The data were normalised at a distance of 10 m. The values were recorded at a height of 0.66 m and an angular position of 0° for the radioactive waste cask and 180° for the spent fuel cask. As expected, because the source is not a point source and because of the scattered neutrons, the relation between the dose rate and the distance is not in $1/d^2$ but rather in $1/d$. This is in agreement with the findings in Ref. (4).

Figure 7 shows the variation with the angular position of the neutron and gamma ambient dose equivalents. The values were recorded every 45° around the cask, 1 m away from its surface and at a height of 0.66 m. The asymmetry of the neutron dose rate around the radioactive waste cask (higher values between 90 and 225°) is due to the presence of the second cask (for the measurement point at 180°) and to the scattering in the concrete wall of the building (at 90°) according to the situation shown in Figure 1.

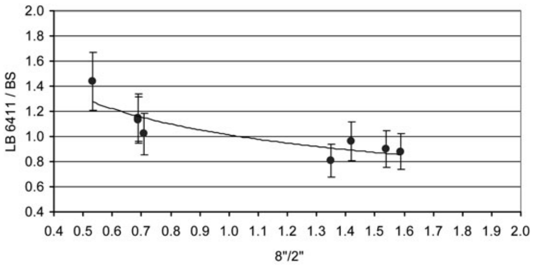

Figure 5. Variation of the ratio of the indications of the LB 6411 monitor and the BS system with the 8- to 2-inch ratio.

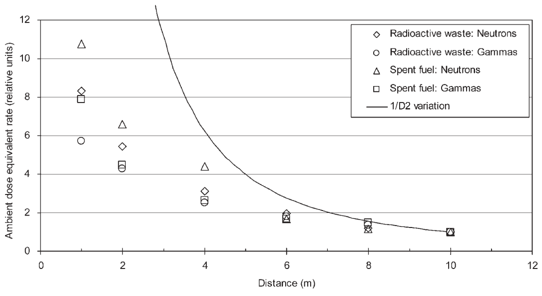

Figure 6. Variation with the distance from the surface of the cask of $\dot{H}^*_n$ (10) and $\dot{H}^*_\gamma$ (10) at 0.66-m height and an angular position of 0° for the radioactive waste cask and 180° for the spent fuel cask. Values have been normalised to 10 m.

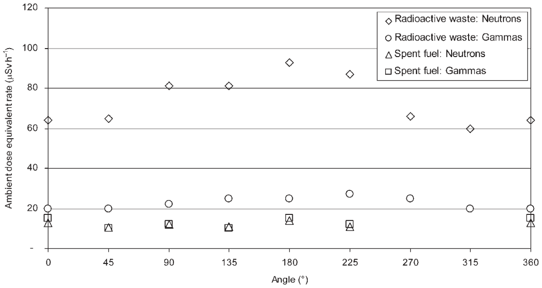

Figure 7. Variation with the angular position of the ambient dose equivalent rate at a distance of 1 m from the surface of the cask and 0.66-m height, measured with an LB 6411 neutron monitor and an Automess Szintomat 6134A gamma monitor.

CONCLUSIONS

A complete characterisation of the neutron field was performed in the vicinity of a spent fuel cask and a high-activity radioactive waste cask at the Swiss intermediate waste and spent fuel storage facility. The neutron spectra were determined with the Bonner sphere spectrometer, allowing for accurate ambient dose equivalent determinations. The neutron field around the cask containing vitrified radioactive waste was found to be harder and more intense than the one around the cask containing spent fuel, resulting in much higher ambient dose equivalents around the former. The spectra at two different heights along the side wall and on the top of the vitrified radioactive waste cask are very similar. For the spent fuel cask a slight difference of shape can be seen between the spectra at two different heights explainable by the burn-up distribution of the spent fuel. A more pronounced difference appears on the top of the cask due to the difference of shielding. The neutron ambient dose equivalent measured with the LB 6411 neutron monitor are in good agreement with those obtained using the Bonner spheres (agreement within 20%), except for large distances away from the cask (e.g. 10 m) where the neutron spectrum is soft and for which the LB 6411 neutron monitor overestimates the values by almost 50%. This shows that this type of instrument, widely used for routine measurements inside the storage facility, provides sufficiently accurate or conservative results. It is an important confirmation for area monitoring while the room is being progressively filled with new containers.

ACKNOWLEDGEMENT

The authors are grateful to the collaborators of the ZWILAG facility for their kind assistance and to Dr. Franz Cartier from the HSK for his collaboration in the campaign.

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