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CALIBRATION AND VALIDATION OF A WIDE RANGE SEGMENTED GAMMA RAY SCANNING INSTRUMENT FOR THE MEASUREMENT OF LOW AND INTERMEDIATE LEVEL WASTE

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ABSTRACT

This paper describes the design and calibration of a Wide Range Segmented Gamma ray Scanning (WR-SGS) assay instrument for the measurement of both Low and Intermediate Level Waste (LLW and ILW) in 200 litre drums. The instrument employs a single shielded and collimated high purity germanium (HPGe) detector to quantify the radionuclide content of the waste. One of the novel features of the instrument is the use of an automated variable aperture collimator, which allows the vertical segment height to be adjusted, and allows the scanning of intermediate level waste drums, with significant surface dose rates. Conventional SGS measurements may be performed where the drum is rotated and measured in vertical segments. Alternatively, faster measurement can be made using continuous helical scanning of the drum as it rotates. A gamma ray emitting transmission source is used to correct for waste density. In place of a conventional shutter, the shielded transmission source is moved to a shielded storage position to eliminate background radiation arising from the transmission source. Using this approach, higher activity transmission sources may be used in order to achieve adequate density corrections for higher density drums.

These new features makes the WR-SGS suitable for the measurement of drums containing exempt level waste to intermediate level waste, as well as drums with a wide range of density. Results will be presented from the calibration of the instrument using horizontally displaced line sources to simulate distributed sources and the results will be compared with benchmarked MCNP Monte Carlo calculations.

INTRODUCTION

The WR-SGS has been designed to extend the range of applicability of the SGS radioactive waste assay technique. The conventional SGS minimum detectable activity (MDA) is often determined by leakage arising from the transmission source and the resulting gamma ray background, especially if a high activity transmission source is used for the measurement of higher density drums. The upper range of measurable drum activity is also limited by high dead time at higher activity levels. Finally, fixed segment-scanning means that during much of the measurement time the HPGe detector is moving but not acquiring data as it is moved into position to count the next segment.

In order to overcome these limitations and extend the range, three novel features have been incorporated into the WR-SGS. The first is a shielded and collimated strong transmission source (typical activity 30 mCi) which, when not in use, is placed in a shielded storage safe such that gamma rays from the source cannot be seen by the HPGe detector. The second is a variable aperture collimator with an automated aperture that can be varied under software control to reduce HPGe detector dead time when high activity drums are being measured. It should be noted that the calibration changes when the aperture changes so that more calibrations are required. The third innovation is the use of continuous helical scanning in place of fixed segment scanning. The present paper describes the calibration and validation using MCNP modelling [1] of a WR-SGS incorporating these features, which has been installed and is in operation in a laboratory of the Australian Nuclear Science and Technology Organisation (ANSTO).

Figure 1 Photograph of the ANTECH WR-SGS installed at ANSTO.

SYSTEM CALIBRATION AND SIMULATION MODEL BENCHMARK

For a specific collimator opening the WR-SGS is calibrated with a single scan of a reference source, which is placed centrally in a void drum. Either line or point sources may be used for the calibration, line sources being preferable for their analogy to volume sources (along the vertical axis). At ANSTO Eu-152 line sources were used for the system calibration. Note that the calibration is done for a void drum: there is no need to repeat calibration for various matrices.

For the simulation benchmarking an Eu-152 point source, placed centrally inside a void drum, was preferred for its low attenuation of the gamma rays. The source, which consists of a 1 mm diameter bead embedded in a PMMA (Acrylic) plastic disk (diameter 25.4, thickness 3.5 mm), is oriented with its axis horizontal. During a scan the drum is rotated, resulting in an average thickness of the plastic of 3.9 mm. The gamma ray attenuation ranges from 7% to 3% at energies of respectively 122 and 1408 keV. In the simulations the source is modelled as a small PMMA sphere with a diameter of 3.9 mm and the photons are started uniformly inside a sphere of 1 mm diameter.

The simulation model is benchmarked to the experimental data from the central segment (#7) of the scan. Where possible the model used in the Monte Carlo simulations is based on design and manufacturing dimensions and material descriptions. In this aspect the “Quality Assurance Data Sheet” [2] with exact crystal dimensions for the detector. However, in order to obtain good agreement between simulated and measured detection efficiencies, especially for the lower energy range, it was necessary to adjust the thickness of the dead layer of the crystal.

The best agreement between simulation and experiment was obtained with a dead layer thickness of 1.18 mm, which is considerably larger than the nominal value of 0.7 mm from the data sheet. The general agreement between simulation and experiment is very good, as can be seen in Figure 2. The curve with the simulation results is based on calculated values for equidistant (10 keV) photon energies, while the curve for the experimental results is based on quadratic fits to data points at the energies of the major Eu-152 peaks. The quadratic fit of the natural logarithm of the efficiency to the natural logarithm of the energy is commonly used for the efficiency calibration of HPGe detectors [3]. It requires the energy range to be split in two: above and below the (user selected) knee energy of 350 keV. The (emission) grab or data acquisition time was 1000 s. The collimator opening was 70 mm. In this paper the error bars in the figures and the reported errors in the tables correspond to 95% confidence interval (2 σ).

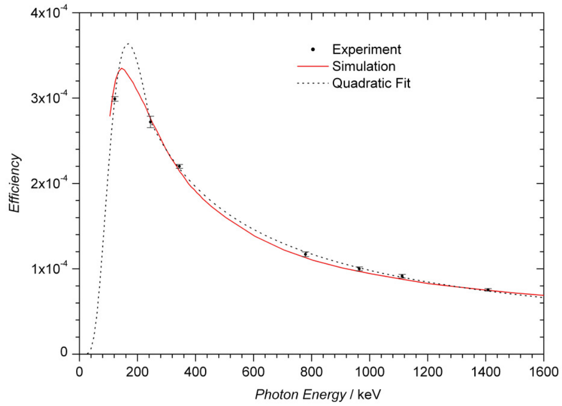

Figure 2 Experimental and simulated detection efficiencies as a function of energy.

Figure 3 shows the excellent agreement between simulation and experiment for a 13 segment scan in which an Eu-152 point source is placed centrally inside an empty drum. The Variable aperture collimator is set to the maximum opening of 70 mm. The small deviations cancel out and can be explained by a small vertical positioning difference between simulation and experiment.

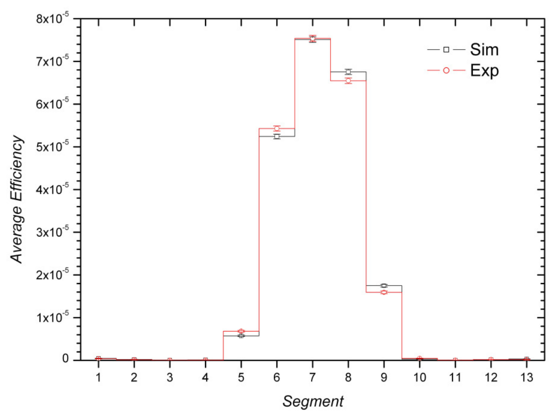

Figure 3 Experimental and simulated average detection efficiencies (at 1408 keV) for an Eu-152 point source placed centrally inside a void drum.

For purposes of the simulation, and in order to mimic the movement of the detector head during the (helical) scan the detection efficiency of each segment is calculated by averaging the simulation results for the detection efficiency at 8 vertical positions ranging from the bottom to the top of each segment. The experimental results are based on a grab time of 1000 s for each segment.

The Variable Aperture Collimator (VAC) provides a means of reducing HPGe detector dead time when measuring high activity drums or sources and extends the range of activities that can be measured by the SGS technique. The use of the VAC and other features of the WR-SGS are described in detail, elsewhere [4]. It must be noted that the calibration must be repeated for each collimator aperture that will be used to measure waste. Currently the recommended apertures are 3.5, 14 and 70 mm. In routine operation data from the drum pre-scan using the Geiger-Muller dose-rate probe and drum weight data from the internal load cell are used to select the appropriate collimator aperture from a user defined look-up table. Typically different collimator apertures may be used for the transmission and emission measurements of the same drum. This feature provides great flexibility in operation and allows the WR-SGS to be automated for a wide range of measurement situations.

LINE SOURCES

Once the system is calibrated it is ready to measure drums with a wide range of matrices and densities that comply to the SGS standard: relative uniformly distributed activity and (per segment) uniformity of the matrix. The production of a series of verification drums satisfying the above requirements is inconvenient and expensive. Instead, drums are used with a uniform matrix and a number of source insertion tubes in which (encapsulated) sources can be inserted. The verification drums employed by ANSTO have 6 source insertion tubes, which are arranged following a spiral from the centre to the edge of the drum (see Table 1 and Figure 4). The design is intended to approximate a uniform homogeneous activity throughout the drum when the drum is rotated and measured. The source insertion tubes are made of PVC plastic, with a 12 mm inner diameter and a 15.9 mm outer diameter.

Table 1 Polar coordinates of the position of sources inside the drum.

Source #	R (mm)	Angle (°)
1	62	43
2	141	331
3	180	261
4	212	221
5	242	171
6	270	90

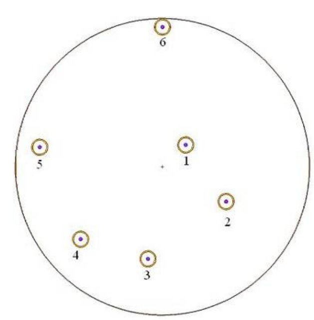

Figure 4 MCNP geometry plot with a vertical section of the void drum.

In the vertical direction the uniform distribution of activity is readily achieved by utilising line sources: the detection efficiency for the segments is nearly constant as can be seen in Figures 5 and 6. As expected the top and bottom segments have a significantly smaller response.

The results for the measurement of one Eu-152 line source placed centrally in a void drum (SAT9) and that for 6 Eu-152 line sources placed inside a drum with foam matrix (average of measurements F000001_04 and F000001_05) are shown in Figure 5, while Figure 6 shows the results for 6 Eu-152 line sources placed inside a drum with wood matrix (average of measurements F000003_11-13) or with sand matrix (average of measurements F000004_12-17). The efficiency values are based on the 1408 keV peak. The scan conditions were: 8 segments, 70 mm collimator opening, 24 mm pedestal height, 810 mm scan height.

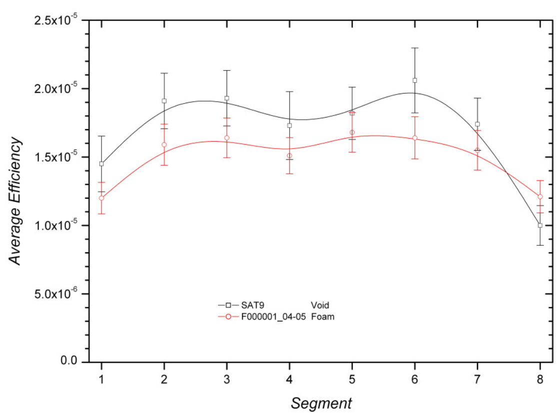

Figure 5 Experimental average detection efficiencies for Eu-152 line sources placed in the centre of a void drum.

The net masses of, respectively, the foam, wood and sand matrices, which are measured with the weigh scale embedded in the WR-SGS, are respectively 6.9, 158.0 and 332.6 kg. The net matrix volume is 2.12×10^5 cm^3 (based on an inner height of 829 mm and a inner diameter of 572 mm), which results in densities of 0.0325, 0.745 and 1.57 $g{\cdot}cm^{-3}$ for foam, wood and sand matrices respectively.

The simulation results for the measurements with the Eu-152 line sources are generally in good agreement with the experimental results as can be seen in Table 2. The significantly higher, i.e. with a deviation larger than the corresponding relative error, simulation result for the foam matrix may be attributed to the simplification of the model: due to the very light density (and the lack on the exact material composition) the matrix was simulated as void. Except for the measurement with ID SAT9, where a single source was placed at the centre of a void drum, 6 sources were inserted in the drum at positions described in Table 1 and Figure 3. In the column labelled M the matrix of the drum is given: Void, Foam, Wood and Sand are respectively denoted by V, F, W and S. To reduce the statistical error for the experimental results the values from repetitive scans and from segments 2 to 7 were averaged. The scan conditions were: 8 segments, 70 mm collimator opening, 24 mm pedestal height, 810 mm scan height. Per segment (emission) grab times were 100 s, except 600 s for SAT9 and 200 s for F000004_15-17.

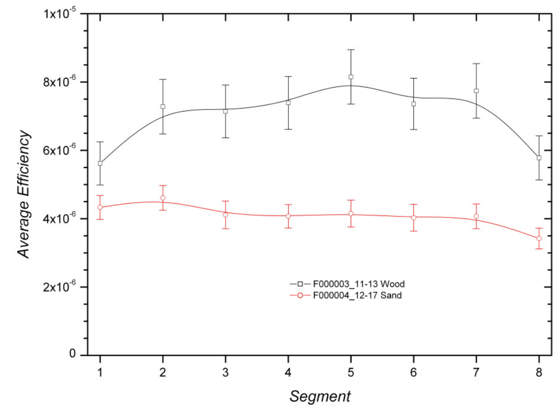

Figure 6 Experimental average detection efficiencies for 6 Eu-152 line sources placed inside a drum with wood or sand matrix.

Table 2 Central segment average detection efficiency (1408 keV) for Eu-152 line sources placed in various matrices.

Measure-ment ID	M	Average Efficiency					
		Simulation		Experiment		Sim/Exp	
		Value	Error	Value	Error	Value	Err.
SAT9	V	1.78E-05	2.41E-07	1.86E-05	8.70E-07	0.96	0.05
F000001_ 04-05	F	1.74E-05	1.07E-07	1.60E-05	5.92E-07	1.09	0.04
F000003_ 11-13	W	7.55E-06	7.08E-08	7.51E-06	3.20E-07	1.00	0.04
F000004_ 12-17	S	4.35E-06	5.45E-08	4.17E-06	1.54E-07	1.04	0.04

POINT SOURCES

To validate the instrument at various conditions two point sources were used: a higher activity Co-60 source (3.07E+07 Bq on measurement date) and a lower activity Cs-137 source (7.17E+05 Bq on measurement date). The higher activity source was measured with a reduced collimator height of 14 mm, while the lower activity source was measured with the maximum collimator opening of 70 mm. The sources were placed centrally in an empty test drum with a single source insertion tube. This insertion tube has a larger diameter than the previous ones in order to allow the insertion of the PMMA disk sources with a diameter of 25.4 mm. The same drum was used for all measurements: it was filled with the sand for high density matrix measurements.

The net mass for the empty drum and sand matrix were 2.5 and 310.9 kg, respectively. The weight of 2.5 kg of the empty drum is due to the source holder. The difference between the masses of the drums with the sand matrix can be explained by a partial filling of the drum with the single source insertion tube. In fact, as can be seen in Figure 7 the transmission scan results (based on averaging the results from measurements A000011_17-20) for segments 2 to 8 yield an average value of 0.72%, while that of the top segment is 77.6%, which corresponds to a fill height of the drum of 73.5 cm. The calculated density of the sand is equal to 1.64 g·cm^{-3}.

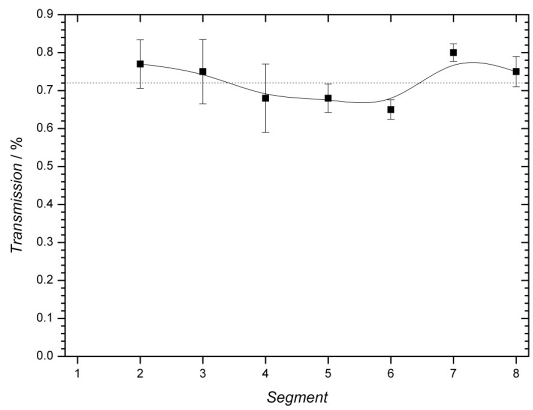

Figure 7 Experimental transmission for 1408 keV photons through a drum with sand matrix. The dotted line gives the average value (segments 2 to 8).

The simulation results for the Co-60 source are 9% higher than the experimental value (Table 3). The deviation being the same for both the empty drum and sand matrix may be explained by the higher attenuation of the source container of this higher activity source. The stainless steel double encapsulation has a wall thickness of 1.3 mm, which results in a transmission of 0.948 for the 1333 keV gamma rays (where we used a mass attenuation coefficient for iron of 0.0520 cm^2·g^{-1} [5]). In the simulation the source was modelled as a PMMA disk source to compare it with the calibration measurement. In fact the stainless steel encapsulation reduces the apparent activity of the source. With the apparent activity of the source the experimental efficiency would be increased by a factor of 1.054. Consequently, the deviation between simulated and experimental results is reduced to 3%, which is in the same order of the errors. The scan conditions were: 8 segments, 24 mm pedestal height, 810 mm scan. Per segment (emission) grab times were 100 s, except 180 s for A000011_25-27.

The simulated result for Cs-137 point source is slightly higher than the experimental value for the foam matrix, whereas for the sand matrix the difference is 18%. Unfortunately, the measurement suffers from poor counting statistics due to the higher attenuation of the sand matrix for the 662 keV photons and the lower activity of the source. Nevertheless, the simulation result is at least 10% higher than the experimental one. This time there is no correction for the source self-attenuation because a PMMA disk source is used in both the simulation and experiment. However, a possible explanation could come from a higher than average density of the sand in the central part of the drum. Taking a closer look at Figure 7, we see that the transmission for segments 4 to 6 is significantly lower (average transmission 0.67%) than that for the other segments (except of course the top segment). In Figure 8 it can be seen that segments 4 and 5 give the largest contribution to the average scan result, and that the discrepancy between simulation and experiment is the largest for those segments.

Table 3 Complete scan average detection efficiencies for point sources placed centrally in drums with various matrices.

Measure-ment ID	Nuclide	M	Average Efficiency					
			Simulation		Experiment		Sim/Exp	
			Value	Error	Value	Error	Value	Err.
A000011_ 25-27	Co-60	F	2.42E-06	4.69E-08	2.22E-06	9.14E-09	1.09	0.02
A000011_ 17-18	Co-60	S	2.11E-07	6.93E-09	1.94E-07	4.44E-09	1.09	0.04
A000011_ 22-24	Cs-137	F	3.01E-05	1.65E-07	2.96E-05	2.98E-07	1.02	0.01
A000011_ 19-20	Cs-137	S	1.03E-06	2.17E-08	8.72E-07	6.76E-08	1.18	0.08

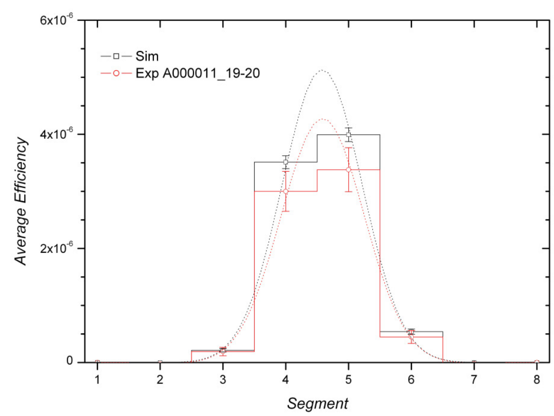

Figure 8 Simulated and experimental average detection efficiencies for Cs-137 point source placed centrally inside a drum with sand matrix.

In the simulation a uniform density of the sand is assumed. An average 0.67% lower transmission of the central segments with respect to the overall average value, does not seem significant, however, one must take into account that the reported values are for 1408 keV photons from Eu-152. At the lower energy of the Cs-137 photons the effect is much larger even if we consider that the photons from the centrally placed source travel across half the distance (one radius) compared to those in the transmission measurement (one diameter). The larger error in the comparison of measurement and simulation for the Cs-137 measurement in sand can be readily explained.

VOLUME SOURCE EQUIVALENCE AND VALIDATION OF MEASUREMENTS

The SGS methodology is based on the assumptions of relatively homogeneous distribution of activity and homogeneity of the matrix inside each segment. Consequently, the measured activity of point sources is likely to be biased. The direction of the bias will be dependent on the matrix and the radial position of the source. In order to compare the reported activities by the GammaScan software with the reference activities found in the source certificates a correction factor is necessary to compensate for the biasing.

Having shown the quality of the simulation model in the previous sections, we will now use the simulations to calculate the correction factor. The results of the simulations are given in Table 4. As expected the correction factor increases with the attenuation of the gamma rays in the matrix. The largest correction factor is for the Cs-137 point source and sand matrix. The calculated efficiencies for Cs-137, Co-60 and Eu-152 are based on peak energies of, respectively, 662, 1333 and 1408 keV. The point source (P) and the single line source (1L) are placed centrally inside the drum; placement of the 6 line sources (6L) is according to Figure 3. The collimator height (H) is given in mm.

Table 4 Simulated average detection efficiencies for point, line and volume sources for drums with various matrices.

H	Nuclide	Src	M	Average Efficiency					
				Point or Line Sources		Volume Source		CF	
				Value	Error	Value	Error	Value	Rel. Err. %
14	Co-60	P	V	2.42E-06	4.69E-08	2.30E-06	3.84E-08	0.95	2.6
14	Co-60	P	S	2.11E-07	6.93E-09	5.21E-07	1.83E-08	2.47	4.8
70	Cs-137	P	V	3.01E-05	1.65E-07	3.04E-05	1.56E-07	1.01	0.8
70	Cs-137	P	S	1.03E-06	2.17E-08	5.13E-06	6.43E-08	4.98	2.5
70	Eu-152	1L	V	1.78E-05	2.41E-07	1.79E-05	1.19E-07	1.00	1.5
70	Eu-152	6L	V	1.74E-05	1.07E-07	1.79E-05	1.19E-07	1.03	0.9
70	Eu-152	6L	W	7.55E-06	7.08E-08	7.52E-06	7.80E-08	1.00	1.4
70	Eu-152	6L	S	4.35E-06	5.45E-08	4.32E-06	5.88E-08	0.99	1.8

The test drums with the 6 line sources provide excellent representations of volume sources: the correction factor is practically equal to unity. Moreover, the observed count rate fluctuations while the drum is being rotated are moderate. Even for the worst-case matrix, sand filled drum, the fluctuations are within 50% of the average as can be seen in Figure 9.

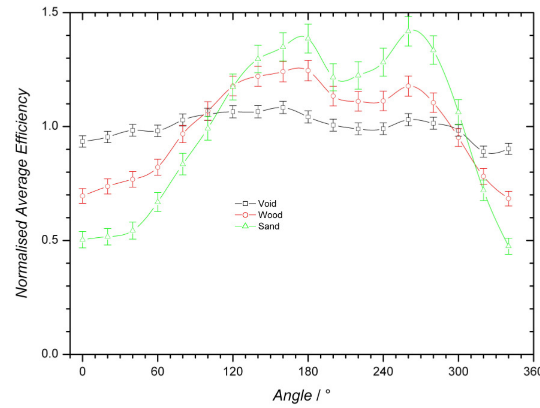

Figure 9 Normalised average efficiency (1408 keV) as a function of angle for 6 Eu-152 line sources with equal activity placed in drums with various matrices. The normalisation for each curve is with respect to the result obtained by averaging over all angles.

The measured activities in Table 5 are obtained by applying the correction factors (in the CF column) to the reported activities by the GammaScan software. In the column labelled M the matrix of the drum is given: Void, Wood and Sand are respectively denoted by V, W and S. The point source (P) and the single line source (1L) are placed centrally inside the drum; placement of the 6 line sources (6L) is according to Figure 3. The collimator height (H) is given in mm. The correction factors are based on simulation results (see table 4) and they compensate for the use of point and line sources as a replacement of volume sources. There is a good agreement with the reference activities: the deviations are within 10%, except those for the sand matrix, which are within 11% (this may be attributed to a lack of uniformity of the sand matrix).

As we have already seen the transmission for the sand matrix is 0.72% for 1408 keV photons, which is reduced to 0.57% for photons with an energy of 778 keV. From the point of view of the modelling a small change in the matrix density will result in a significant change in detection efficiency. Experimentally, even with a strong transmission source the number of counts for a typical segment grab time of 100 s are limited, and hence the statistical error will limit the precision of the matrix correction factor used in the calculations according to the SGS standard. Finally, it must be noted that the stated uncertainty in the various source certificates is equal to 3% and which has not been included in the reported error calculations.

Table 5 Comparison between measured and reference activities for various sources and matrices.

Measure-ment ID	H	Nuc.	Src	M	CF	Measured Activity (Bq)		Ref. Activity (Bq)	Meas. / Ref.	Rel Err %
						Value	Error			
A000011_ 25-27	14	Co-60	P	V	0.950	2.93E+07	4.51E+04	2.82E+07	1.039	2.6
A000011_ 17-18	14	Co-60	P	S	2.469	2.63E+07	2.29E+05	2.82E+07	0.934	4.9
SAT6	70	Cs-137	P	V	1.010	6.99E+05	8.54E+03	7.21E+05	0.969	1.4
A000011_ 22-24	70	Cs-137	P	V	1.010	7.46E+05	3.72E+03	7.17E+05	1.041	0.9
A000011_ 19-20	70	Cs-137	P	S	4.981	6.39E+05	2.30E+04	7.17E+05	0.891	4.4
SAT9	70	Eu-152	1L	V	1.004	1.45E+05	1.94E+03	1.43E+05	1.015	2.0
F000001_ 04-05	70	Eu-152	6L	V	1.029	7.81E+05	4.86E+03	8.40E+05	0.930	1.1
F000003_ 11-13	70	Eu-152	6L	W	0.996	7.66E+05	6.58E+03	8.39E+05	0.913	1.6
F000004_ 12-17	70	Eu-152	6L	S	0.992	7.54E+05	5.53E+03	8.44E+05	0.894	2.0

CONCLUSIONS

After the benchmarking procedure, the simulation model correctly predicted the detection efficiency for a variety of nuclides and matrices, increasing considerably the credibility of the calculated point and line source to volume source correction factors. These factors are necessary to correct the activities reported by the instrument for comparison with reference values. After the applying the corrections, we report a general agreement between reference and reported values within 10%, validating both the robustness of the system's hardware and software for a broad spectrum of photon energies and matrices.

Moreover, it is also demonstrated that the test drum design employed by ANSTO with 6 Eu-152 line sources placed along a spiral path provide an excellent equivalent of a volume source.

These measurements and their comparison confirm not only the validation of the calibration process but also the correct functioning of both the hardware and software of the WR-SGS for the measurement of radioactive waste.

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