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Comparison exercise on activity determination of radioactive waste drums in Taiwan

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HIGHLIGHTS

- Comparison exercise on activity determination of radioactive waste drums in Taiwan was performed.
- Radioactive waste drums with uniform and non-uniform radioactivity distribution were prepared at NRSL.
- Testing drums were measured by 7 laboratories using high resolution gamma-ray spectrometry.
- E_n and B_i values were used as parameters to evaluate the comparison results.
- Highest discrepancies were found for drums with the rod-shaped source located in the center of the drum.

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ABSTRACT

The National Radiation Standard Laboratory of Taiwan organized in 2014 a comparison exercise by distributing 210 L drum-typed samples to seven radioactive waste analysis laboratories in Taiwan. Four drums were filled with uniformly distributed active carbon, water, resin and concrete, respectively and five drums were filled with cracked metals and heterogeneously distributed radioactive sources. Measurement uncertainties of participants results are in the range 3–40% ($k=2$) and about 96% of the reported results produced E_n values (ISO, 1997) smaller than one for drums with activity uniformly distributed. The minimum discrepancies, expressed as B_i values (ISO, 1997), of drums with heterogeneously distributed ^{137}Cs and ^{60}Co were 0.34 and 0.17, respectively.

1. Introduction

Radioactive waste produced by nuclear power plants or nuclear facilities is usually classified into two categories, according to the activity level. For activity levels below the clearance level, the waste can be processed and disposed as non-radioactive material. For activity levels above the clearance level, the waste shall be put into dedicated storage facilities (IAEA, 2004). In particular, the clearance limit for ^{60}Co or ^{137}Cs is 100 Bq/kg.

Several laboratories have already organized comparison exercises on activity measurements of radioactive waste in drums. For instance, Dean (2009) employed a radioactive-adsorbed resin to make a reference drum to be used as sample for the comparison exercise. In 2010 and 2012 the National Radiation Standard Laboratory (NRSL) of Taiwan organized two comparison exercises (Yuan et al., 2010; Yeh et al., 2012) on activity measurements of 210 L waste reference drum filled with a radioactive solution of known activity. For both studies, the activity in the drums was uniformly distributed and good comparison results were shown.

As continuation of previous comparisons organized by NRSL, in the present study four testing drums containing homogeneously distributed radioactive materials and five testing drums containing non-homogeneously distributed radioactive materials were prepared at NRSL as comparison samples. Seven laboratories in Taiwan participated in this comparison exercise. All drums were circulated to participants for measurement and the final results were analyzed by NRSL.

2. Method

Participating laboratories were coded using letters from A to G. Three types of analytical equipment were used by participating laboratories. In particular, four laboratories used Canberra ISOCS system, one laboratory used Canberra Q2 system, and the remaining two laboratories used ORTEC ISO-CAR system (Canberra Inc., 2009a, 2009b, 2004, METEK/ORTEC, 2012). Noteworthy is that

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Canberra Q2 system employs three HPGe detectors within a 10 cm thick lead shielding while the other systems employ only one HPGe detector without shielding. Six laboratories were equipped with turntables to rotate the drums while the other one moved the detector manually to measure the drums by different directions. The analysis software applied were Genius 2000 for Canberra systems (Canberra Inc., 2009b) and Isotopic for the ORTEC system (METEK/ORTEC, 2008).

In this work, all testing drums had the same size. The wall thickness, height and diameter were 2 mm, 86 cm and 56 cm respectively, and they were classified into two types. Type I drums contained ^{137}Cs and ^{60}Co homogeneously distributed in different matrices (active carbon, water, resin and concrete), for a total of four drums. To check for homogeneity degree of radioactivity in type I drums, the surface dose rate was measured at NRSL in four directions (0°, 90°, 180°, 270°) around the drums by using a scintillation detector (ATOMTEX survey meter/AT1121). For each direction, measurements were performed at different heights with intervals of 15 cm from top to bottom of the drum. The dose rate differences were less than 25%. Such differences are considered compatible with the measurement result uncertainty of method used for dose rate estimation. The activity of each drum was unknown in advance except for the drum filled with water. This water solution was prepared using a reference source traceable to primary standards of NRSL. The solution-filled drum hosted 360 PVC cans (9 layers of 40 cans). Each can was filled with 450 mL of inactive diluent into which 50 mg of the standard solution (^{137}Cs or ^{60}Co) were added by electronic dispenser. Twenty of these cans were randomly selected to check activity concentration and homogeneity (Yuan et al., 2010). The homogeneity test showed a difference of activity concentration between cans of about 18% for ^{137}Cs and 16% for ^{60}Co. As a whole, 398 kBq of ^{137}Cs and 350 kBq of ^{60}Co were added in the cans, and these two activity levels were approximately twenty times higher than the clearance limit.

Type II drums contained radioactive material with known activities which were not evenly-distributed. As shown in Figs. 1 and 2, slice-shaped sources or rod-shaped sources were used to fill the drum with fragments of metal tubes. The slice-shaped sources containing 89 kBq of ^{137}Cs and 33 kBq of ^{60}Co had 40 cm diameter and were located at the upper 1/6, middle and lower 1/6 part of the drum, respectively. There was only one slice-shaped source in each drum. The rod-shaped sources were located at the quadrant of the drum or in the center of the drum. Diameter and length of the rod-shaped sources were 1.5 cm and 60 cm respectively and contained 87 kBq of ^{137}Cs and 78 kBq of ^{60}Co. There was only one rod-shaped source in each drum. The radioactivity content of each type II drum was traceable to the national standards of NRSL with an uncertainty less than 1.6% (k=2).

All analytical equipment and related analysis software used by participants in the comparison exercise were also available at NRSL. According to analysis software calculation approach, counting efficiencies for gamma photons are computed assuming that the testing object has a uniform radioactivity distribution. In order to take into account of source inhomogeneity and apply proper geometry corrections to the counting efficiency, some modeling studies should be performed to simulate the real geometry (i.e. hot spot distribution in the drum, dimension and material of the drum, source size). In the present work, one participating laboratory performed such modeling studies and the corresponding B_i values obtained after simulation are closer to 1 for most of geometries considered (Table 1).

In the planning phase of this comparison exercise, a meeting with participants was held at NRSL to present and discuss the whole process including description of radioactivity distribution in each drum. After words, all the testing drums were distributed to participants and each participant had two days to measure them.

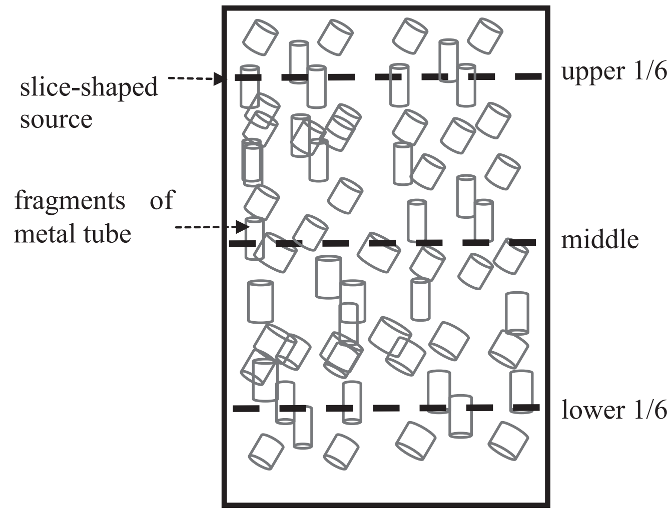

Fig. 1. Slice-shaped source position in the testing drum.

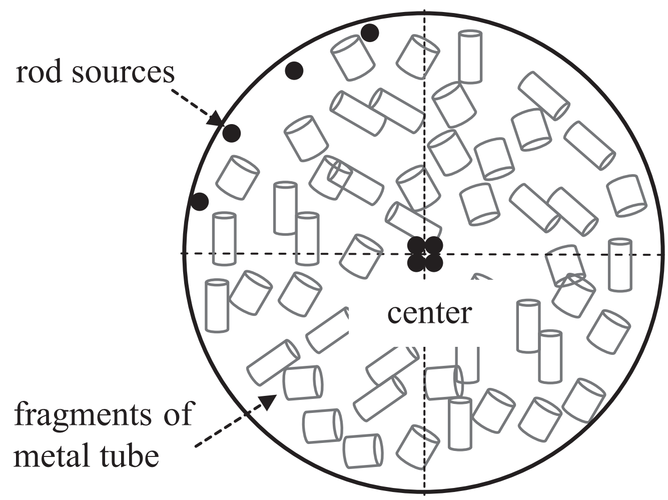

Fig. 2. Rod-shaped source position in the testing drum.

Table 1
Results of the geometry simulation study performed by one participating laboratory to take into account the source inhomogeneity.

Site	Nuclide	B_i value	
		After simulation	Before simulation
Upper 1/6	^{60}Co	1.2	1.3
	^{137}Cs	1.1	1.1
Middle	^{60}Co	0.8	0.9
	^{137}Cs	0.8	0.9
Lower 1/6	^{60}Co	0.8	0.6
	^{137}Cs	0.9	0.6
Center	^{60}Co	1.1	0.7
	^{137}Cs	0.9	0.6
Quarter	^{60}Co	1.1	1.1
	^{137}Cs	0.9	0.8

The reference values of the comparison exercise and related uncertainties were obtained as follows:

(a) For type II drums and for the type I drum filled with water solution, the reference values were determined by calculation, according to the certified activity value of the reference standard solution and taking into account dilutions performed. The associated uncertainties were determined as combined

Table 2
Reference value of each drum in this work. Uncertainties are expressed as expanded uncertainties at $k=2$.

Category	Drum ID	Material in the drum	Activity (kBq)	
			^{60}Co	^{137}Cs
I	1	Solution	350.2 ± 3.5	398.5 ± 6.0
I	2	Concrete	109.1 ± 8.1	119 ± 11
I	3	Resin	266 ± 23	27.7 ± 4.2
I	4	Active carbon	128 ± 19	$13{,}500 \pm 1300$
II	5	Upper 1/6	33.1 ± 0.5	89.3 ± 1.0
II	6	Middle	33.1 ± 0.5	89.3 ± 1.0
II	7	Lower 1/6	33.1 ± 0.5	89.3 ± 1.0
II	8	Quarter of the drum	77.78 ± 0.93	86.7 ± 1.4
II	9	Center	77.78 ± 0.93	86.7 ± 1.4

standard uncertainties following the law of propagation of uncertainties, with a coverage factor $k=2$ (JCGM, 2008).

(b) For type I drums filled with concrete, resin and active carbon, the reference values were determined as arithmetic mean of participants results. The associated uncertainties were determined as standard deviation of the mean of participants results, with a coverage factor $k=2$.

The reference values for each drum are reported in Table 2. Reference values for the last two type II drums (rod-shaped source) were different from the first three type II drums (slice-shaped source), as two standard solutions were used.

The E_n value and B_i value described in Eqs. (1) and (2) were used as parameters to evaluate the comparison results. Measurement results with $E_n \leq 1$ were considered acceptable (ISO, 1997).

$$E_n = \frac{M_{lab} - M_{ref}}{\sqrt{U_{lab}^2 + U_{ref}^2}} \tag{1}$$

$$B_i = \frac{M_{lab}}{M_{ref}} \tag{2}$$

where:

M_{lab} is the value reported by each participant;
M_{ref} is the reference value;
U_{lab} is the expanded uncertainty of the reported value;
U_{ref} is the expanded uncertainty of the reference value.

3. Results and discussion

Both for type I and type II drums, no information about uncertainty components considered in the final uncertainty budget of reported results was provided by participants. This lack of information is reflected in the discussion and interpretation of results, in particular for type II drums, where inhomogeneity of radioactivity distribution should represent a main uncertainty component of counting efficiency.

3.1. Type I drums

The E_n and B_i values are shown in Fig. 3. The expanded uncertainty of activity values reported by participants ranges from 18% to 40%, reaching the highest values for laboratory D in the case of drums filled with concrete and resin.

About 96% of measurement results came with E_n values below 1, this showing a good performance of participating laboratories. Only two measurement results from laboratories D and E respectively showed E_n values higher than 1. The two outlier laboratories were asked to double-check their results and the large discrepancy with respect to the reference value was confirmed. The B_i value was 0.66 and 1.36 for the two laboratories respectively.

3.2. Type II drums

Due to the strong effect of source inhomogeneity on measurement results and to unrealistically low uncertainties provided by participants (ranging from 3% to 34%), the E_n and B_i values, shown in Fig. 4, were calculated at NRSL taking into account an additional uncertainty component related to source inhomogeneity. Such uncertainty component was estimated assuming a rectangular probability distribution to describe source inhomogeneity and was combined in quadrature with the measurement result uncertainty provided by participants. Discrepancies, expressed as B_i values, between reported results and reference values were in the range 0.17–1.46. The maximum differences between measured values and the reference value for 7 laboratories were shown for the rod-shaped source located in the center of the drums. This geometry is in fact characterized by higher self-shielding effect and proper correction for self-attenuation should be applied. It is worthy to mention that one laboratory (code E) among the seven laboratories showed a better performance, with E_n values in the range 0.098–3.924 and 0.034–2.726, for ^{137}Cs and ^{60}Co results respectively.

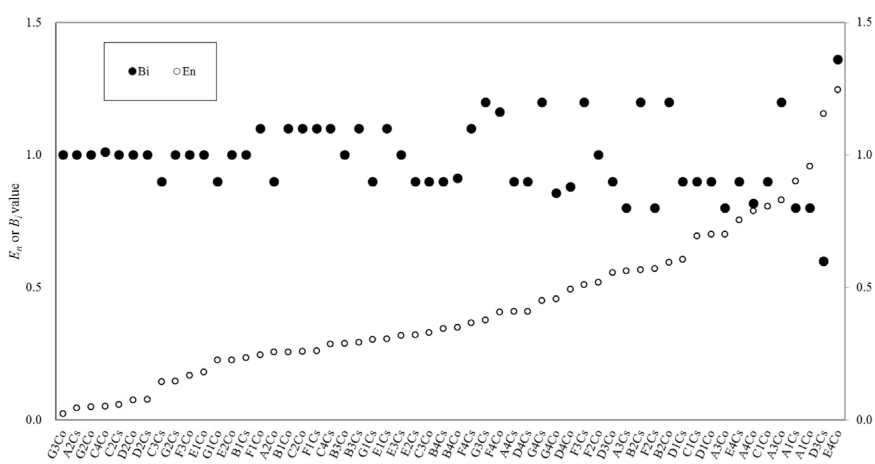

Fig. 3. Results of all participants expressed as E_n and B_i values for type I drums and sorted by the E_n value.

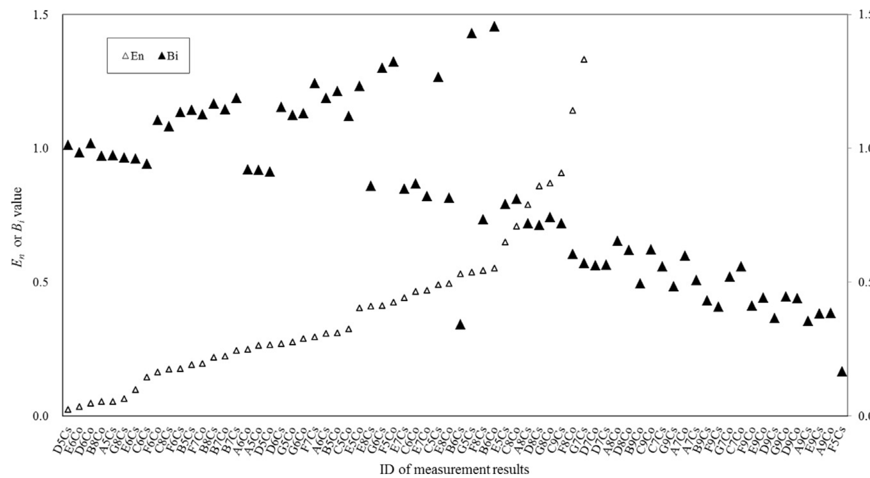

Fig. 4. Results of all participants expressed as E_n and B_i values for type II drums and sorted by the E_n value.

Considering the additional uncertainty component due to source inhomogeneity, the expanded uncertainty of activity values reported by participants ranges from 32% to 67%, reaching the highest value for laboratory F. About 36% of reported results for type II drums had E_n values greater than one. These results are characterized not only by larger discrepancy with respect to the reference values, but also by uncertainties lower than the ones of results with E_n values less than one. In this case, an underestimation of uncertainty values and/or an incomplete uncertainty budget can be considered as a main reason affecting the laboratory performance.

4. Conclusions

In the present work, a comparison exercise was performed using two types of drums. For type I drums, about 96% of reported results were acceptable. For type II drums, mainly due to the source self-shielding effect and heterogeneity, a higher discrepancy between reported results and the reference values was shown, the maximum difference occurring when the rod-shaped source was located in the center of the drum. The minimum discrepancies (B_i values) for type II drums were 0.34 and 0.17, for ^{137}Cs and ^{60}Co respectively. When taking into account the uncertainty component due to source inhomogeneity, expanded uncertainties of measurement results of participants are higher for type II drums, this confirming the importance of properly estimating such uncertainty component in the case where proper geometry correction to the counting efficiency is not applied. In the case where such correction is, instead, applied using experimental or computational approaches, the uncertainty component due to the application of the correction should be included in the final uncertainty budget.

From this study, it gathered the necessity of properly surveying the radioactive waste before putting it into the 210 L drums to avoid hot spots in certain parts of the drum. It is recommendable to preliminary measure the surface dose rate of the drum to detect potential heterogeneity and take this into account when performing gamma-ray spectrometry measurements. That would increase measurement accuracy and quality of analytical results, thus preventing from erroneous judgment of radioactive waste classification and improving radiation protection measures. Through participation in such kind of comparison exercises, laboratories in charge of radioactive waste management can assess their technical capability and eventually improve their performance. Due to specific criticalities encountered by laboratories who participated in this kind of comparisons organized by NRSL over the later years, a further need of similar comparisons and proficiency tests has been identified. Information from participating laboratories regarding uncertainty components estimation and specific measurement procedures applied is a main asset for proper interpretation of comparison results and better design of future tests.

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