EXPERIENCE IN THE DEVELOPMENT AND OPERATION OF FACILITIES FOR PRIMARY CHARACTERIZATION OF WASTE FROM NUCLEAR FACILITIES

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The article focuses on methodological and hardware support of radioactive waste characterization, including the one associated with radiation characteristics of materials measured to support the decision-making on its release from regulatory control. The study provides an assessment of uncertainties and their influence on the measurement results. It presents the experience of the Czech company JSC VF in the development of automated installations allowing to address this problem with some examples of such installations operating in various countries provided.

Keywords: *radioactive waste, release from regulatory control, JSC VF, plastic scintillators, scaling factors, key radionuclides, difficult-to-measure radionuclides, radionuclide vector, decommissioning, measurement geometry.*

Introduction

Waste segregation according to the radiological hazard level, including waste release from regulatory control is seen as a basis providing effective radioactive waste (RW) management during operation and decommissioning of nuclear facilities (NF). Fully automated installations, the so-called FRM (FreeRelease-Measurement), manufactured by the Czech company JSC VF (VF a.s.) are nowadays widely used to address this problem. JSC VF has been manufacturing such equipment for almost 20 years, and with the development of measuring equipment and legal framework in various countries to which this equipment has already been supplied, its designs have been gradually improved. FRM measurement systems are only part of the process enabling the release of materials from regulatory control with their design depending on a number of factors. The article aims to single out the factors governing the requirements for the FRM device itself. For example, it relates to the container size in which the measurements are implemented, the maximum weight of a material batch, the selected technology given a specific type of measured materials, opportunities for hybrid approach application, etc. These factors are important since the requirements for the equipment and pre-measurement preparation of material, scaling factors identification and relevant uncertainties, other uncertainties, for example, those related to the non-uniformity of the package filling, the non-uniformity of radionuclide activity distribution over the material, etc. directly depend on these factors. The article covers all stages of the measurement process from pre-measurement preparation of material to quality assurance and service personnel training. The latter topic has been considered in the context of the software system for FRM installations with WAMIS (Waste Activity Management Information System) being viewed as its integral part. In conclusion, the paper analyzes the operating experience associated with specific FRM installations manufactured by VF.

Radioactive waste characterization and the process of material release from regulatory control

In terms of RW management safety and its increased efficiency, RW segregation according to the radiological hazard level is seen as a most important stage of the process. Considered essential are the actions on the release of material inventories from regulatory control providing significant reduction in the volume of material categorized as radioactive waste and requiring specific measures for its processing and disposal. Provisions of the national legal framework regulating this area, which may differ from country to country, are viewed as a basis for these actions. This article does not intend to analyze these requirements, but only stresses some commonly ignored context and theoretical area of possible exceptions under the process of material release from regulatory control (the so-called conditional release).

Specific activities of certain categories of radionuclides are viewed as criteria for such release. These are based on the generally accepted dose criterion of 10 μSv/year [1, 8, 9, 11, 17].

Using various conceptual and mathematical models, which were not considered in this paper, general levels for the release of materials have been specified. These are expressed in terms of the specific activity of each radionuclide or a group of radionuclides. For example, for typical key (reference) radionuclides ^{60}Co and ^{137}Cs with each one of them producing different effect on people when released into the environment (it differs by 2—3 times), the same limit usually amounting to 100 Bq/kg has been established [4, 10]. Although other less stringent limits were set in the past amounting to 300, 400 Bq/kg [3, 5, 6, 10, 12].

Already at this level, certain conservatism, which is not taken into account any further, can be observed: the overwhelming majority of methods providing material release from regulatory control are based precisely and solely on generally established limits, for example, 100 Bq/kg for ^{60}Co, ^{137}Cs, etc. However, the same conceptual and mathematical models suggest no control over the future use of the released materials. If they are used, for example, in the construction of access roads in the form of crushed stone, railway embankments, etc. then the levels of such conditional release can be 10—30 times higher than the levels established for the release of further uncontrolled material (Figure 1), thus, allowing to save some free space in low-level waste repositories.

Adherence to the following condition (rule of sums) is seen as a criterion (factor) suggesting that

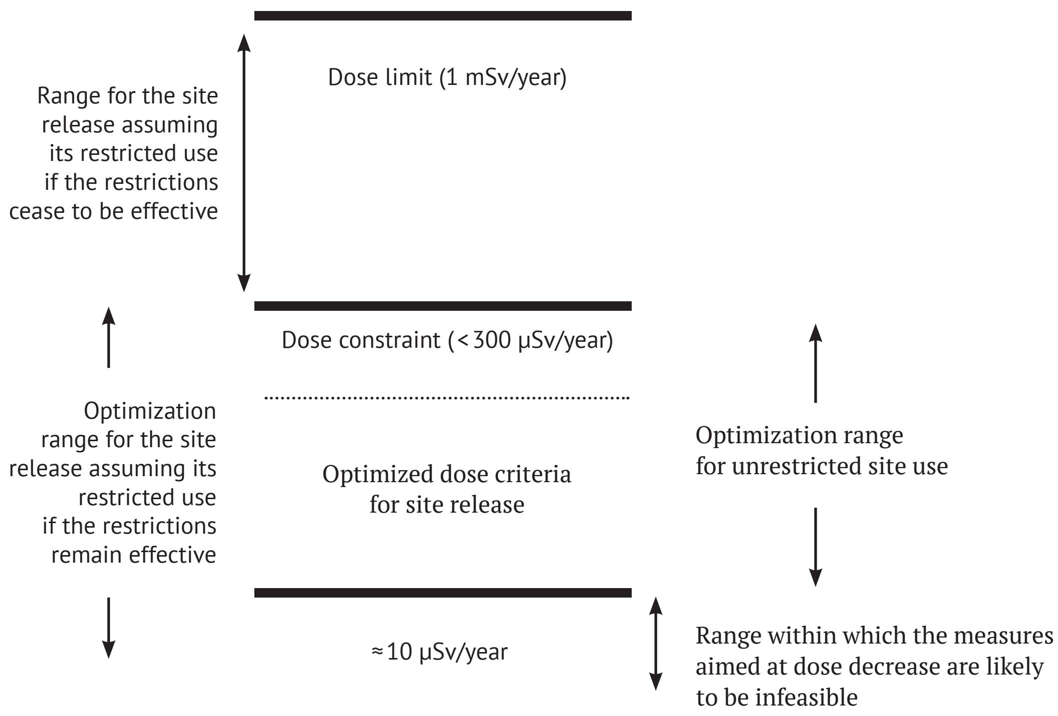

Figure 1. Optimization of dose constraints and effective dose ranges for a critical group of population during site release from regulatory control [1]

a batch of radioactive material (RMB) can be released from regulatory control:

$$S + \Delta S \leqslant 1, \qquad (1)$$

$$S = \sum_{i=1}^{n} \frac{C_i}{C_i^{clr}}, \qquad (2)$$

$$\Delta S = \sqrt{\sum_{i=1}^{n} \left(\frac{C_i}{C_i^{clr}} \right)^2}, \qquad (3)$$

where S is the sum of ratios between the specific activities of the monitored radionuclides and the release level, the preliminary list of which is specified for the RMB;

ΔS is the right border of a one-sided confidence interval S;

i is a conditional serial number of the monitored radionuclide;

C_i is the measured specific activity of the i-th radionuclide in the RMB;

C_i^{clr} is the release level (specific activity) of the i-th radionuclide in the RMB as specified in regulatory documents;

ΔC_i is the right border of a one-sided confidence interval for the measured specific activity of the i-th radionuclide depending on a number of factors including statistic uncertainty, calibration tolerance of the measurement path, scaling factors for difficult-to-measure radionuclides, etc.

n is the number of monitored radionuclides preliminary identified for the RMB.

It should be noted that all components of the total uncertainty in the previously proposed expressions should correspond to the same confidence level and the measurement result itself cannot be recognized as reliable without proper assessment of its uncertainty in the specified confidence interval.

Key Provisions to be addressed during the establishment of systems for RM characterization and release from regulatory control

Radioactive material characterization provides for some provisions that should be considered and adopted to allow for their successful application. These may include the following:

Regulatory averaging

Material release limits differ slightly from country to country and are more or less similar to the same levels (e. g., 0.1 Bq/g for ^{60}Co, ^{137}Cs). However, quite often, averaging opportunities may differ. Measurement tools are basically capable of determining the overall activity of a container or a waste package. Its average specific activity can be calculated after the weight of an empty container and the one filled with radioactive material is identified, the net weight of the radioactive material is calculated and its activity is measured. Measurement uncertainty in case of weighing is usually less than 1%. Definitely, it can be considered as the least critical parameter for the overall measurement uncertainty. Obviously, at different points of the package, material activity can vary from the average one. However, different countries have quite different requirements for the maximum possible averaging value. Basically, it ranges from 1 to 300—1,000 kg, in some exceptional cases averaging up to 5,000 kg is allowed.

Limiting the size and volume of the measuring container

The second limitation refers to the size of the measured package. Often this parameter may vary in a quite wide range, although, usually measured are standard 200-liter drums used for material storage. Nevertheless, there are no requirements for a specific geometry, and it is the operator, sometimes even the equipment manufacturer, who must demonstrate the feasibility of the adopted engineering solutions to the regulatory body.

The choice of measurement geometry is essential. Self-shielding effect of measured material being an objective physical phenomenon manifests itself in case of any measurement method. Its manifestation appears to be more pronounced at higher densities of the measured material or rather its bulk density. For mineral wool or plastic, this effect is not quite pronounced (up to about 1,000 kg/m^3), but in case of concrete chips (1,000—2,000 kg/m^3) or metal waste (up to 7,800 kg/m^3) it appears to be quite significant.

The greatest efficiency is achieved when the shape of the measuring container is as far as possible similar to the "sphere", i. e., the smallest ratio between its volume and surface area. However, in practice, cube-shaped containers are most commonly used in case of systems with a 4π-sensor layout geometry. In the case of “frame” systems, when the measuring container with the material passes through a frame fitted with detectors, the cross-section shape is similar to a square (surface/perimeter ratio is minimum). However, this factor ultimately turns out to be a disadvantage even though this solution may first seem somewhat logical and beneficial in case of its optimization. In practice, a 5—10 cm layer of a material subject to measurements statistically produces 10—100 times greater influence on the measurement result than the underneath layers located deeper in the container.

Therefore, this factor should be accounted for in the applied measurement technique. In this case, various approaches can be applied:

1) a very conservative approach is introduced limiting the permissible volumetric weight, whereas no advantage is taken of the design capacity capabilities of the installation;
2) a conservative constant is introduced into the measured specific activity due to which the acceptance value (the measured activity at which the waste can be released from regulatory control) is reduced, thus, increasing the volume of material that cannot be released from regulatory control;
3) stricter requirements for the pre-measurement material preparation are introduced, as well as for the characterization of each measuring container fragment providing high uniformity of radioactive material distribution in the container resulting in somewhat increased requirements for the pre-measurement stage.

An alternative approach can be applied as well. For example, this challenge can be evaluated at the initial stage already with a decision made on the application of a "thin" measurement geometry, i. e., geometry in which the distance between the detectors is set so that the flow of material passing along the detectors is as "thin" as possible even if the production capacity of such system may prove to be somewhat inefficient. Conversely, if the plant operator is able to ensure unambiguous uniformity (homogeneity) of the measured material in the container (for example, in case of concrete homogenization by crushing and stirring), then such material can be effectively measured using high-performance equipment with a "deep" container.

Influence of this parameter on measurement uncertainty usually amounts to over 10%, but even 100% uncertainty in complex geometric configurations cannot be excluded.

Measurement of difficult-to-measure radionuclides

Difficult-to-measure radionuclides (DTM-RN) are primarily alpha- and beta-emitters, but these may also involve some gamma-active radionuclides emitting low-energy radiation, such as ^{241}Am. This paper provides no detailed information on relevant measurement technique. Another point seems interesting to us, namely, under what circumstances the difficult-to-measure radionuclides (DTM-RN) may provide a significant contribution to the criterion established for waste release from regulatory control and when it will only affect its uncertainty. It is even more important to understand what should be the number of DTM-RNs generally included into list of measured radionuclides when a decision is made on whether the material should be released from regulatory control or not. The radionuclides contained in the measured material can be conditionally divided into key (reference) (KEY-RN) and difficult-to-measure radionuclides (DTM-RN) mathematically related to KEY-RN factors. In the case of spectrometry, the latter ones are called scaling factors (SF), whereas in the case of total gamma activity measurements (Total Gamma) these are radionuclide vectors (RNV) [2, 6].

A reasonable boundary should be identified by establishing a balance between cost and time savings and a conservative approach in determining the list of such radionuclides. No matter of what equipment is used, it will never have a zero minimum detectable activity (MDA). Therefore, during any material measurement, the minimum reliably measured activity is equal to MDA. If an "infinite" list of DTM-RN radionuclides that may affect the release criterion is assumed, then by multiplying the MDA by the "infinite" number of SFs assumed for individual theoretical DTM-RNs and summing the resulting values, a release criterion being greater than 1 can be derived for a material being considered "clean". The impact produced by this parameter on the measurement uncertainty usually amounts to over 5—10%. However, 50% uncertainty may be the case if it's considered impossible to accurately determine the SF.

Selecting the type of measurement

The topics related to the decision making on the selection of proper measurement systems (with plastic scintillators, NaI (Tl) scintillation crystals, HPGe technology or a combination of these methods) will be generally discussed at the end of this article. There are other types of purpose-designed scintillation detectors, for example allowing to detect neutrons in mixed fields, as well as analysis devices, for example NGA-01 [16, 7]. However, these are rarely applied to provide the material release from regulatory control and therefore are not discussed in this paper.

The measurement uncertainty associated with the impact produced by a specific type of a detecting device usually amounts to 1—5%. It should be emphasized that the factors governing the choice of a particular geometry and the challenges associated with DTM-RN problem produce a more significant influence than the activity measurement itself which depends on the selected physical measurement principle.

Quality assurance

Quality of a measuring device largely depends on the accuracy and stability of the measuring method. WAMIS software (WAMIS software), i. e., the superior software system of the FRM installation, provides functions requiring regular daily, monthly, yearly checks implying the use of control point sources, typical phantoms of the measured material, as well as regular checks of the weighing system, etc.

Measurement records are stored in a database, since the checks performed are seen as a condition providing the validity of actual measurements.

In addition to metrological quality assurance, WAMIS/FRM also provides maximum possible protection against errors caused by the human factor, i. e., actions of the service personnel. Users and operators log on to the system through secure accounts and the operator's identity is captured in summary protocols.

Conformity checks performed for mass measurements, measurements performed to assess the degree and uniformity of container filling, the non-uniformity of activity distribution within the measured container or container segment are seen as an integral part of the measurement process.

Methodical approach to optimal technology selection

Measurements performed for characterization purposes and to provide the release of radioactive materials from regulatory control involve specific procedures related to specific equipment, measurement principle, shape and size of a container, properties of the released material, as well as the environmental conditions in which the process takes place (for example, an unusually high gamma background is observed in the Chernobyl zone mainly associated with ^{137}Cs; this fact has a significant impact on the measurement itself).

Irrespective of indirect non-destructive measurement methods applied, the choice of the measurement principle is primarily influenced by the measured material, its theoretical radionuclide characteristic, the size of the fragments, the size of the measuring container used for its storage. In addition, this principle is also influenced by the uniformity of the measuring container filling with the material, the uniformity of the material itself and, no less important, the uniformity of the contaminant distribution in its volume.

Usually, measuring equipment can automatically check only some part of the above properties, and even a lesser part of it can be compensated through calculations. For example, a procedure specified to measure the weight of a container together with its known volume and monitored filling along its height can provide the measuring system with data on the average bulk density of the material. However, all other parameters or their uncertainty will be compensated by conservative inclusion of relevant factors into the measurement result uncertainty. The only way to keep these uncertainties to minimum is to select an optimal measurement geometry and a method to be applied during pre-measurement material preparation.

However, even frequently mentioned spectrometric systems based on semiconductor germanium detectors (HPGe) cannot measure the DTM-RN content, and uncertainties similar to those caused by the uncertainties in the scaling factor identification will be present similarly to the systems based on plastic scintillation detectors (Total Gamma), i. e., radionuclide vectors (RNV) [2, 6].

The table below summarizes possible methodological approach to be applied during the decision-making on the optimal measurement technology.

Methodological approach to be applied during the decision-making on the optimal measurement technology

1 2 3

	1	2	3	4
Key restrictions, segregation	Does not contain naturally occurring radionuclides at all or contains only some minor amount of such radionuclides	Contains significant amounts of naturally occurring radionuclides	Contains a lot of various key radionuclides which cannot be safely segregated into batches	Hybrid solutions
Technology	Scintillation plastic detectors, geometry similar to 4π	Scintillation detectors NaI (Tl), measurement geometry implying material passage through a frame	HPGe detectors, measurement geometry implying material passage through a frame	FRM-02 + FRM-03
FRM manufactured by JSC VF	FRM-02, FRM-02B	FRM-06	FRM-03	FRM-02C
Typical application	Release of metal waste, construction rubble with constant specific activity by ^{40}K	Release of construction waste	Release of material containing earlier unknown key radionuclides	Released is not only metal waste, but also construction waste after ^{40}K measurement and statistical control of key radionuclides

Continuation of table

Unambiguous advantages of the method	The highest installation performance-cost ratio. Measurements can be followed up even in case of multiple detector failure	High measurement rate, possibility of ^{40}K and key radionuclide (^{60}Co,^{137}Cs) differentiation	Spectrometric measurement of all detectable gamma emitting radionuclides allowing for direct DTM conversion using SF	Advantages are similar to those of the FRM-02 system (see p. 1) also allowing for the detection of significant deviations from given radionuclide composition
Disadvantages	If the key radionuclide is unknown, higher degree of conservatism is applied, as well as in case of higher concentration of ^{40}K and other naturally occurring radionuclides	The measurement path should be stabilized. Even in case of only one detector failure, failure of the entire device occurs	Detectors should be cooled which complicates the technology. Even in case of only one detector failure, failure of the entire device occurs. High cost of the device	Statistical control strongly depends on the homogeneity of the measured material and significantly lengthens the measurement process. High cost of the device
Approximate capacity	4-8 t/h	10 t/h	0.5-2 t/h	10 t/h

To select an optimal technology, selection algorithm is recommended to be applied by moving along the table from left to right. Method 1 is considered as a default one. If its application appears to be impossible due to restrictions, then Method 2 is applied. If there is no further correspondence, then Method 3 is applied with Method 4 being viewed as an alternative option.

Experience of FRM application

First **FRM-02** model (NPP Kozloduy, Bulgaria) [19] designed by JSC VF first appeared at the market in 2004-2006 (Figure 2) [13]. Its designs provided for only plastic scintillation detectors accommodated within a geometry very similar to 4π. A total of 32 detectors were fitted on six sides around the container with the material. Each of them had a volume of 6 dm^3. The detectors provided for a 48 % coverage as regards the inner surface of the chamber, whereas 53 % coverage was provided as regards the spatial angle of the detectors relevant to the center of the chamber (this parameter is believed to be very similar to 4π). The measuring chamber was shielded with 5 cm-thick lead blocks. The function of release factor calculation for a given radionuclide vector (RNV) and deduction of background readings caused by naturally occurring radionuclides, especially ^{40}K has been provided for in this model already. In addition, this unit was equipped with a potential discontinuity detection function, the so-called hot-spot detection.

Figure 2. FRM-02, Kozloduy NPP, Bulgaria

Retrofitted **FRM-02B** unit [13] was supplied to the Ignalina NPP (Lithuania) (Figures 3, 4) [20]. The

Figure 3. FRM-02B, Ignalina NPP, Lithuania

Figure 4. FRM-02B, Ignalina NPP, Lithuania

retrofitting was primarily aimed at reducing the measurement time and thereby increasing the system performance. In practice, it was found that actual measurement time (maximum 3 minutes, usually 30—60 seconds per 1 m^3/1,000 kg of waste) accounted only for some small part of the total cycle time, which also included container loading on the installation with a forklift, conveyor assisted container transfer to the measurement chamber and vice versa. Therefore, the conveyor was equipped with two T-shaped loading positions. This design provides a real-time average measurement cycle of less than 8 minutes, which has been demonstrated during practical tests (conservatively declared minimum capacity was taken equal to 6 t/h).

The next modification of this unit **FRM-02C** [13] was supplied to the Jaslovské Bohunice NPP (Slovakia) (Figure 5). As compared to the **FRM-02B** designs, the main difference accounted for its additional fitting with a high-purity germanium (HPGe) semiconductor detector located on top allowing to:

- check gross deviations from the declared radionuclide vector (RNV);
- perform statistical tests, i. e., to extend the selected measurements, in accordance with the methodology, providing more accurate estimates regarding RVN correctness.

Figure 5. FRM-02C, Jaslovské Bohunice NPP, Slovakia

Further, the design was retrofitted in a way allowing the device to distinguish between the clean zone and the controlled access zone: it's a walk-through device containing two shielded gates. The conveyor and actuators of the screening gates inside the measurement chamber have been retrofitted.

These design modifications allowed to increase the minimum **FRM-02C** capacity to 10 t/h. Thus, it can be considered as a most high-performance model among the installations with a geometry similar to 4π.

In addition to FRM-02C, **FRM-06** [14] and **FRM-24** [15] units have been supplied to Jaslovské Bohunice NPP (Figures 6, 7).

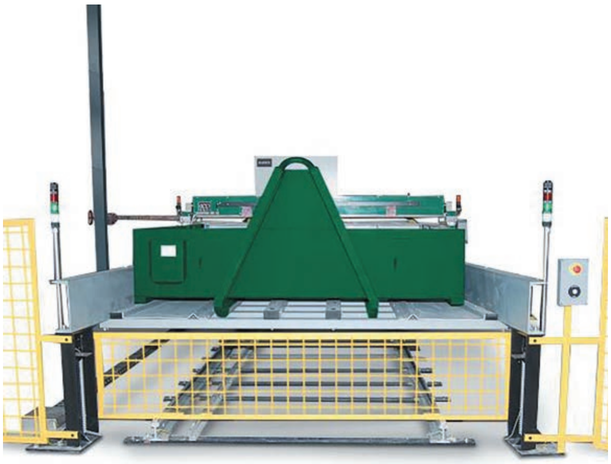

Figure 6. FRM-06, Jaslovské Bohunice NPP, Slovakia

Figure 7. FRM-24, NPP Jaslovské Bohunice, Slovakia

FRM-06 (Jaslovské Bohunice NPP, Slovakia) is designed for spectrometric measurements. It applies 6 large-volume NaI(Tl)-based scintillation detectors anchored to the measurement frame: 3 at the top and 3 at the bottom. They cover a "thin" 3300 × 2000 × 500 mm measurement container. This method represents a compromise between high-cost spectrometry based on semiconductor germanium detectors and high-efficiency plastic scintillators.

At the same time, this design allows not only to decide whether the material in the container is contaminated with key (reference) radionuclide ^{60}Co or ^{137}Cs, but also to provide correct measurements of naturally occurring radionuclides in terms of their specific activity, for example, the specific activity of ^{40}K. As it comes to methods based solely on plastic scintillators, the ^{40}K correction is addressed by multiplying the values from the database by the net waste mass, which seems to be a correct approach, nevertheless, resulting in a higher uncertainty.

FRM-24 [15] (Jaslovské Bohunice NPP, Slovakia) designed for a relatively small amount of waste is worth noting as well. The technology combines large-volume plastic scintillators and large-volume NaI(Tl) detectors built into the construction of a walk-through measurement tunnel.

FRM-03 unit is the newest unit produced by JSC VF [18, 21], which has been successfully put into trial operation at Chernobyl NPP (Figure 8).

Figure 8. FRM-03, Chernobyl NPP, Ukraine

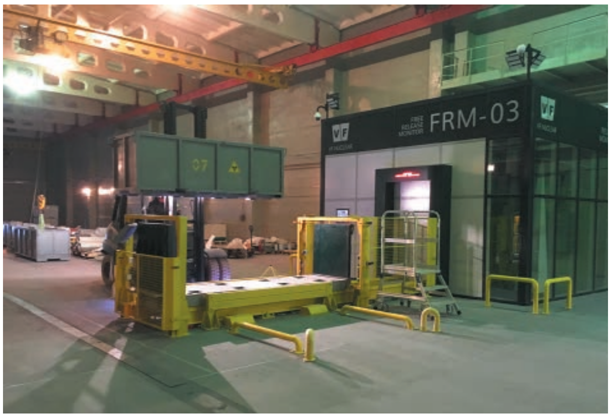

Figure 9. FRM-03, Chernobyl NPP, Ukraine. Container loading with a forklift

The installation has a measuring container with a capacity of 3 m^3 and a total weight (gross) of up to 5,000 kg (Figures 8, 9). The container is repeatedly passed through the measurement frame equipped with three semiconductor germanium detectors with 40% relative efficiency (Figure 10). Such a large container size, on the one hand, helps to reduce the time for waste preparation (fragmentation), on the other hand, it imposes more stringent requirements on the measurement method and material preparation for its release from regulatory control. Practice shows that the greatest efficiency can be achieved when all the factors in general are accurately taken into account.

Figure 10. FRM-03, Chernobyl NPP, Ukraine. Measurement frame with semiconductor germanium detectors in a shielded tunnel made of low background material

Conclusion

This article describes the main types of automatic equipment used in the process of material release from regulatory control in standardized measuring containers. This paper gives no consideration to the options of so-called in situ measurements of large-sized objects being considered as much more time-consuming and complex and providing for strict requirements to the equipment and the personnel engaged in its maintenance.

The article, on the one hand, superficially summarizes the challenges and problems arising in the process of material release from regulatory control, and on the other hand, indicates the associated difficulties in general. It summarizes and overviews the general approaches, types of technologies and key recommendations.

Factors governing the decision-making on the optimal engineering solutions to be applied during measurement waste control have been evaluated: it was noted that not only the type of equipment appears to be essential, but also other factors, the key of which is the measurement geometry, including the container size.

JSC VF is able to offer a solution to any customers and, if necessary, to account for regulatory and methodological requirements established in a particular country. It is one of a few manufacturers that thanks to its know-how and many years of experience is able not only to design, develop and manufacture equipment, but can also provide consulting assistance in the development of measurement flowcharts and procedures providing material release from regulatory control.

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