THEORETICAL, INSTRUMENTAL, AND OPERATIONAL CONSIDERATIONS IN CLEARANCE OF SLIGHTLY RADIOACTIVE MATERIAL – AN AUSTRIAN PERSPECTIVE

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ABSTRACT

A new radiation protection law designed to implement the recommendations of the 29/96/EURATOM directive has recently been adopted in Austria.

The conditions to be fulfilled prior to clearance derive from the 10 μ Sv/year additional dose concept. Volumetric and surface activity concentration limits for each radionuclide result. In order to assure compliance with the rather restrictive values sophisticated clearance measurements are called for. International experience has shown that acceptable results in terms of quality, throughput, and price, are practically only achievable by using a purpose built automated measurement unit, working on the principle of total- γ counting. Calibration and measurement protocol play a key role in obtaining reliable results. The issue of homogeneity of the activity distribution becomes important when clearance of objects exceeding regulatory mass (surface) limits is attempted.

Experience with installation and operation of a clearance measurement unit by Nuclear Engineering Seibersdorf GmbH are discussed.

INTRODUCTION

Clearance of slightly radioactive material is defined [1] as 'removal of radioactive materials or radioactive objects within authorized practices from any further regulatory control by the regulatory body'.

Clearance is based upon the realization that the radioactivity of some materials is so low as to render them harmless. In other words, the dose associated with exposure to these materials is trivial. The currently recommended value for the trivial dose is 10 - 100 μ Sv/year [2], or 10 μ Sv/year for a single clearance practice [3]. These recommendations have been adopted for use in the EU [4].

The practical application of this concept necessitates the calculation of volumetric and/or surface activity concentration limits for each nuclide from the trivial dose and *vice versa*. The results critically depend on the assumed exposure scenario, as well as on a number of other variables, such as the physical state of the material to be cleared, the disposal pathway, *etc.* However, the accuracy and validity of such calculations are in question, *e.g.*, there are indications that activity

concentration limits thus calculated might be an order of magnitude too conservative [5]. Nevertheless, adherence to the 10 μ Sv/year concept seems to be the rule rather than the exception in Europe. (There are other ways of dealing with clearance, *e.g.*, the case-by-case approach in the USA or the 0,4 Bq/g total activity concentration limit in use in the UK.)

Situation in Austria

A new radiation protection law designed to implement the recommendations of the 29/96/EURATOM directive has recently been adopted in Austria. A new radiation protection ordinance will follow shortly. Hence, a novel legislative framework for clearance has been created. Of special importance for clearance is the transition from the previously used 10 μ Ci/m³ (0,4 Bq/g at a density of 1 g/cm³) total activity concentration limit to the set of nuclide specific activity concentration values, e.g., 0.1 Bq/g for ⁶⁰Co, 0.5 Bq/g for ¹³⁷Cs, and 0.03 Bq/g for ²²⁶Ra.

Nuclear Engineering Seibersdorf GmbH, as the only facility for collection, treatment, conditioning, and interim storage of low and intermediate level waste in Austria, is in the process of adapting its clearance operations accordingly.

Instrumentation

In order to assure compliance with the rather restrictive activity concentration limits sophisticated clearance measurements are called for. These can be performed either manually or automatically and either spectrometrically or by total- γ counting. International experience has shown that the necessary quality and throughput at a justifiable price is practically only achievable by using an automated total- γ counting measurement unit (see Table I).

	•	Throughput	Homogeneity Determination	Price	Automatic Documentation
Manual		low	no	Low	no
Automatic	Spectrometric	medium	yes	High	yes
	Total-γ Counting	high	yes	medium	yes

Table I.	Com	parison	of	Clearance	Measure	ement Methods
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These units typically employ shielded large area plastic scintillator detectors in a 4π geometry. Because of operation in the vicinity of the detection limits, calibration and measurement protocol play a key role in obtaining reliable results. In particular, the effects of background, natural radioactivity of the measured material, and the self-shielding of the measured material have to be considered. Calibration is typically performed with a key nuclide, *e.g.*, ⁶⁰Co, ¹³⁷Cs, yielding a set of calibration factors (one for every detector). Separate calibrations are performed for different types of material, *e.g.*, pipes, sheet metal, insulation, concrete rubble, *etc*. The measured activity is considered 'key nuclide equivalent' and activities of all nuclides of interest, including α - and β -emitters not directly measurable, are evaluated using a nuclide vector. Hence, the reliability of the results hinges on the accuracy of the nuclide vector, which can be measured or estimated based on process knowledge. This means that the method is ideally suited for large clearance waste streams with well-defined and essentially constant composition (NPP decommissioning)

and becomes progressively more labor intensive as the variability of the clearance waste stream composition increases.

Homogeneity

The immediate result of a clearance measurement using an automated total- γ counting measurement unit are the count rates of the *n* detectors, $\{Z_i\}_{i=1,2,...n}$. The calculation of the average nuclide specific activity concentration of the material using the detector calibration factors, the nuclide vector, and the mass (surface) of the material is straightforward. However, the regulatory mass (surface) limit of 300 kg (1000 cm²) presents a problem for measuring material with larger mass (surface). This can be dealt with in a conservative way by using the regulatory mass (surface) limit irrespective of the actual mass (surface) of the material, possibly leading to not clearing 'clearable' material. Hence, a more sophisticated approach relying on the concept of homogeneity of activity concentration distribution deserves closer scrutiny. The calculation of the activity concentration distribution a(x,y,z) in the material from a set of detector count rates:

$$\{Z_i\}_{i=1,\dots,n} [cps] \to a(x, y, z) [Bq/cm^3]$$
(Eq. 1)

is not straightforward as can be appreciated by examining the form of the functional dependence between a(x,y,z) and $\{Z_i\}_{i=1,2,...n}$:

$$Z_{i} = \frac{1}{4\pi} E_{\text{int},i} \iint_{S_{i}V} a(x, y, z) \frac{1}{r^{3}} \overrightarrow{ds} \bullet \overrightarrow{r} dV$$
(Eq. 2)

where E is detector efficiency, S is detector area, V is waste package volume, and r is the distance between the detector and the waste package. While $\{Z_i\}_{i=1,2,...n}$ can easily be calculated from a(x,y,z), the inverse problem is underspecified unless $i \rightarrow \infty$ (infinite number of detectors). In practice, the volume of the material package is subdivided into j hypothetical sections with each section having a constant activity concentration $a_i(x,y,z)$ and the following problem:

$$\{Z_i\}_{i=1,\dots,n}[cps] \rightarrow \{a_j(x,y,z)\}_{j=1,\dots,m}[Bq/cm^3]$$
(Eq. 3)

is then solved numerically using the maximum-likelihood method. Deviations of individual a_j's from an average activity concentration value indicate inhomogeneities in the activity concentration distribution. Important work in this field has been performed at VKTA Rossendorf [6, 7]. The practical application of the homogeneity concept is further complicated by the lack of relevant regulatory limits.

Implementation

A modern total- γ counting clearance measurement unit has recently been installed in Seibersdorf. The background in the building is on the order of 0.1 µSv/h. The unit features 10 plastic scintillator detectors (50 x 50 x 5 cm) in a 4 π geometry. The measurement chamber is shielded by 50 mm of lead from all sides. The background count rate of a single detector is <100 cps. A detection limit of 300 Bq ⁶⁰Co within 60 s is claimed. The unit is intended primarily for handling material in standard 200-L drums but can be used for differently packaged material as well,

including the option to measure long items with the door open. An integral scale with a 1000 kg weight limit is built into the bottom of the measurement chamber.

Materials slated for clearance include excavated soil, concrete rubble, empty 200-L drums, decay waste, etc. A separate calibration was performed for each material type by using identical or similar inactive material with a ⁶⁰Co or a ¹³⁷Cs point source imbedded in various positions. Simulating a homogeneous activity concentration distribution by using point sources leads to an overestimation of the activity concentration by up to 30%. Hence, when possible, calibration drums with homogeneous activity concentration distribution were prepared and used for calibration, e.g., a 100-L soil drum with ca. 2250 Bq/kg of ⁶⁰Co. The natural radioactivity of materials like excavated soil and concrete rubble is so high that a significant increase in detector count rates compared to background was observed with 'inactive' material. The software, expecting a count rate decrease due to self-shielding, was unable to calculate a self-shielding correction. Hence, the ability to correctly account for this effect during actual measurement proved to be essential. The original software of the clearance measurement unit allowed only for nuclide specific entry and subsequent subtraction of the naturally occurring nuclide activity in Bq/g, e.g., ⁴⁰K. In cooperation with the unit manufacturer, a new option has been implemented that allows for an automatic subtraction of the count rate in cps observed with the 'inactive' material without the need to specify the offending nuclides.

To date, approximately 100 t of excavated soil and concrete rubble in 100-L and 200-L drums have been cleared and shipped off site for conventional recycling. Work on calibration and measurement protocol is ongoing.

CONCLUSION

An automated clearance measurement unit, working on the principle of total- γ counting, has been installed by Nuclear Engineering Seibersdorf GmbH and operational experience has been gained in the course of clearing approximately 100 t of material.

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