Proceedings of the 19th International Symposium on the Packaging and Transportation of Radioactive Materials PATRAM 2019 August 4-9, 2019, New Orleans, LA, USA

Establishment of a rapid method for the classification of CERN inter-sites radioactive transport by measurement of dose equivalent rate.

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Abstract

The European Organization for Nuclear Research (CERN) has premises on both sides of the French/Swiss border. As a consequence of the exploitation of the machines the activation of material is unavoidable. Inter-sites transports on public roads between the different sites represent around 1800 packages per year. Therefore, compliance with the ADR (European Agreement on the International Carriage of Dangerous Goods by Road) regulations is required. In order to classify this large number of transports efficiently, a pragmatic methodology to classify CERN's activated radioactive goods via dose equivalent rate measurements has been developed.

The ADR classification for material is based on the activity of the radionuclides contained therein. The purpose of the proposed method is to determine this via a simple dose equivalent rate measurement. In a first step, a study of the relationship between the dose equivalent rate and the specific activity of radionuclides has been carried out for 896 irradiation scenarios modeled with ActiWiz 2ⁱ. This method relates the dose equivalent rate of different materials with their nuclide inventories, taking into account the irradiation scenario and characteristics of the materials considered (chemical nature, mass, dimensions ...). A measurement of the dose equivalent rate of an object for which the irradiation history and its characteristics are known, then allows deducing the activity of the radionuclides it contains. In succession, the use of RAW-IIⁱⁱ allowed for determining dose equivalent rate limits that reflect specific transport classes for objects parametrized by their material type, dimension and irradiation scenario. This allowed for constructing envelope scenarios, removing the dependency on the often unknown irradiation history.

Due to the high volume of transports, a set of various software tools has been developed which automatize the documentation of the complete transport process workflow.

Introduction

CERN's premises consist of several separated sites, all along the CERN accelerator complex located, in France and Switzerland (Figure 1). The two main sites are the Meyrin site and the Prévessin site where the major part of the CERN infrastructure is situated, like offices, service buildings, workshops but also experimental areas and the "low energy" accelerators located at the surface. The smaller sites are the access points to the underground tunnels and caverns of the Super Proton Synchrotron (SPS) and the Large Hadron Collider (LHC). Consequently, CERN's radioactive material transport between CERN sites implies the use of public roads and has to follow ADR.



Figure 1 – Location of the CERN sites along the CERN accelerator complex.

According to ADR, for the transport of dangerous goods the proper UN number has to be assigned in order to define all the required provisions. For Class 7 Radioactive Material this classification is based among others on the

- 1. nuclide inventory of the material,
- 2. dose equivalent rate in different distances from the material and/or the package,
- 3. surface contamination of the material.

The number of CERN's radioactive transports planned for the Long Shutdown number two (LS2) is quite high and is estimated with an average of about twelve per day. The transports have to be done under very tight time constraints as they are part of an efficient repair, maintenance and modification process of CERN's accelerators and experiments. Consequently, the transport classification, i.e. the assignment of the correct UN number has to be done within short time delays.

While the dose equivalent rate and the surface contamination can be measured with hand-held instruments within a reasonable time delay, the identification of the nuclide inventory is more difficult. CERN's radioactive material is mainly produced by activation of accelerator and experimental components during beam operation producing a complex nuclide inventory inside the material. The determination of this nuclide inventory by means of Monte-Carlo simulations, gamma spectroscopy and radiochemical analysis is cumbersome and time consuming. Therefore, in practice these methods are not systematically applicable for CERN's inter-sites transports.

To reach the very ambitious goal of combining ADR requirements with CERN's requirements a new approach for the transport classification is proposed: classification of (non-contaminated) material by dose equivalent rate only. This document will describe the scientific basis for such an approach.

In a first step solid objects of simple geometrical forms, like shielding blocks or simple magnets had been assessed. However, the approach can be extended to a major part of CERN's radioactive material.

Principle

At CERN, most radioactive materials is activated in the radiation fields in and around the accelerators, they contain a more or less complex radionuclide inventory – with respect to the type of radioisotopes and activity. According to the ADR the package classification of the radioactive goods is based on this nuclide inventory and is done using the following equation:

$$\sum_{i} \frac{A_{i}}{T_{i,j}} \le 1 \tag{1}$$

with

A_i Activity or Activity concentration¹ of nuclide i.

Transport limit of nuclide (i) of the package classification (j). This is the activity or activity concentration limit given in the ADR for each nuclide depending on the package classification, e.g. for the classification type A package, other form: $T_{i, \text{ type A}} = A_2(i)$.

For each package classification, the activity based transport limits are given in the ADR. Table 1 lists some package classifications and the corresponding transport limits.

Table 1 – Package classification (j) and corresponding transport limits $T_{i,j}$ based on the total activity or activity concentration of nuclide (i)

Package classifications (j)	Transport limits T _{i,j} per nuclide (i)
A. conc. exempt Material	Activity concentration for exempted material [Bq/g]
A. exempt Consignment	Activity for exempted consignment [Bq]
Excepted Material Package	10 ⁻³ A ₂ [TBq]
LSA-I	30 x Activity concentration for exempted material [Bq/g]
LSA-II	10 ⁻⁴ A ₂ /g [TBq/g]
LSA-III	2 x 10 ⁻³ A ₂ /g [TBq/g]
Type A, other form	A ₂ [TBq]

With the help of the Monte-Carlo code FLUKA^{iii, iv} the nuclide inventory inside a block with given dimensions, material composition and density can be calculated for a given irradiation scenario. The irradiation scenario is defined by four parameters:

- E, the beam energy or momentum.
- P, the position of the material relative to the beam impact point/loss point, taking also into account possible shielding as well as different geometries of the impact point (target geometry).
- T_{irr} , the duration of the irradiation of the material.
- T_{cool}, the cooling time, counted from the end of the irradiation.

If the activity or activity concentration A_i of the nuclide inventory is fulfilling equation (1) for a certain transport limit $T_{i,j}$, the related transport classification would be applicable for the activated material block.

 $^{^{1}}$ For reasons of simplicity, A_{i} is used for both total activity as well as the activity concentration. To which one it is referring is depending on the form of the transport limit $T_{i,j}$ used. Some of them are defined in total activity (e.g. activity limits for type A), the others in relative units (e.g. activity concentration limit for LSA-I).

For each nuclide i, the dose equivalent rate \dot{D}_i is proportional to its activity A_i , taking into account the specific conversion factor for ionizing radiation Γ_i that is corresponding to the respective nuclide.

$$A_{i} = \frac{\dot{D}_{i}}{\Gamma_{i}}$$
 (2)

Using

$$\dot{D}_{total} = \sum_{i} \dot{D}_{i}$$
 and $A_{total} = \sum_{i} A_{i}$ and $\Gamma_{total} = \sum_{i} \Gamma_{i} \times A_{i} / A_{total}$

Equation (2) can be written:

$$A_{i} = \frac{(F_{i} \times \dot{D}_{total})}{\Gamma_{i}} \qquad if \qquad F_{i} = \frac{A_{i}}{A_{total}} \times \frac{\Gamma_{i}}{\Gamma_{total}}$$

so the equation (1) becomes:

$$\sum_{i} \frac{(F_{i} \times \dot{D}_{total})/\Gamma_{i}}{T_{i,j}} \le 1$$
 (3)

With last equation, we can calculate a classification of an activation scenario from the measured dose equivalent rate \dot{D}_{total} .

Using the specific conversion factor for ionizing radiation Γ_i the expected dose equivalent rate \dot{D}_i outside the material block can be estimated for each calculated nuclide of an activity A_i . However, in the literature, these tabulated factors are generally based on point like sources. To obtain more realistic results, the dimension of the object has to be considered taking into account the self-absorption as well as the shape. Therefore, more realistic "activity-to-equivalent dose equivalent rate" conversion factors Γ ' have been generated with FLUKA for different materials and geometries at a distance of 10 cm from the material surface, considering a uniform activation. These conversion factors as well as a collection of nuclide inventories have been collected in a database and are available via the software package Radiological Work Station (RAW-II)ⁱⁱ.

It should be noted that the measured dose equivalent rate can be used for complete classification even if pure alpha/beta emitters have been produced as there are always accompanying dominant gamma emitters to be found.

Methodology

Using the conversion factors, the total dose equivalent rate \dot{D}_{total} in a distance of 10 cm from the material surface can be calculated for the sum of activities A_i of the given nuclide inventory.

$$\dot{D}_{total} = \sum_{i} A_{i} \Gamma'_{i}$$

This dose equivalent rate \dot{D}_{total} reflects the radionuclide content inside a given material block for a chosen irradiation scenario and can be used to define the transport limit, (equation (3) = 1). Thus, inversely the measured dose equivalent rate \dot{D} can be used to determine information on the radionuclide inventory for a block with known material and irradiation parameters. If the measured dose equivalent rate in a distance of 10 cm of the material surface

is smaller or equal to \dot{D}_{total} the nuclide inventory of the block fulfils equation (1) for the given transport limit $T_{i,j}$ and the corresponding package classification is a valid option.

Example I, using one standard ActiWiz (a software package that is based on generic FLUKA simulations.) scenario^v:

- Object: Steel cube

Dimension: 16 x 16 x 16 cm³
 Material: Stainless steel 304L

- Density: 7.8 g/cm³

Transport limit

- T_{i, LSA-I}: Activity concentration limit for LSA-I

Irradiation scenario:

- Particle energy of first beam: Protons 1.4 GeV/c (CERN PS Booster)

- Position (particle impact point): 10 cm lateral distance from target (representing a collimator)

Irradiation time: 1 dayCooling time: 1 week

The simulation gives a dose equivalent rate $\dot{D}_{total} = 24.7~\mu Sv/h$ in a distance of 10 cm from the surface of the cube. If the measured dose equivalent rate in a distance of 10 cm outside this cube is lower (e.g. $20~\mu Sv/h$) or equal to \dot{D}_{total} , the radionuclide inventory of the steel block fulfils equation (1) and allows the use of the package classification LSA-I.

Summarizing: in case the main parameters of an activated material are known (see example above), the radionuclide inventory could be determined by dose equivalent rate measurements and simulations.

In general, however the information on the irradiation scenario is not available. Therefore, the dose equivalent rate values associated with certain transport classes have been calculated for many different scenarios and different material positions.

To calculate the nuclide inventories for different irradiation scenarios ActiWiz 2 has been used.

The irradiation parameters used for these studies are listed in Table 2. The combination of these irradiation parameters results in 896 different irradiation scenarios. The beam energies range from 1.4 GeV to 7 TeV, covering the energies of most of the CERN's accelerators (PS Booster, PS, SPS, LHC); irradiation times and cooling times between 1 day and 20 years; and 8 different positions with respect to the beam impact point have been considered.

This selection of scenarios covers almost all cases of material activation occurring inside accelerator tunnels and experimental areas at CERN that are relevant in terms of transport of LS2's goods. If it should turn out during the using phase of the method that a frequent arising scenario is not covered, the list can be extended.

Table 2 – List of irradiation parameters used for the calculation of the dose equivalent rate values.

Beam Energy	Irradiation Time	Cooling Time	Material position
1.4 GeV	1 day	1 day	beam impact
14 GeV/c	1 week	1 week	within bulky material
400 GeV/c	1 year	1 month	adjacent to bulky material

7 TeV	20 years	4 month	nonth close to the concrete tunnel wall (bulky material)		
		2 years	behind massive concrete shielding		
		5 years	10 cm lateral distance target		
	10 years	close to the concrete tunnel wall (beam on			
		target)			
		20 years			

For the steel cube in example I all the 896 different irradiation scenarios of LSA-I classification and the corresponding 896 dose equivalent rate values \dot{D}_{total} have been calculated. The dose equivalent rate are plotted as histogram in Figure 2.

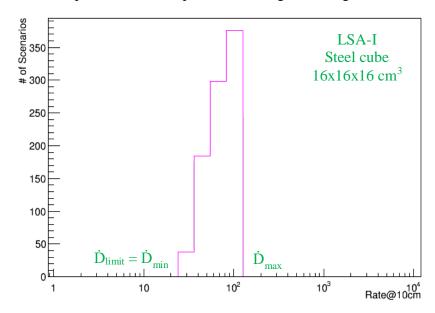


Figure 2 – Histogram of the \dot{D}_{total} values calculated for the 896 scenarios of a steel cube (16x16x16 cm3) and the $T_{i,LSA-I}$. \dot{D}_{limit} refers to the minimum dose equivalent rate.

To classify the material of unknown irradiation history for transport as LSA-1, the lowest dose equivalent rate value \dot{D}_{limit} (envelop) of the 896 values for \dot{D}_{total} , has to be respected.

In the example of the steel block the maximum dose equivalent rate measured in a distance of 10 cm from any surface of the block needs to stay below \dot{D}_{limit} for LSA-I. In this case, the radionuclide inventory of the block fulfils equation (1) with the transport limits of LSA-I no matter if the block was activated for one year next to an impact point in the LHC or it was irradiated for 5 years behind a shielding wall.

In case the measured dose equivalent rate lies between \dot{D}_{limit} and the maximum calculated dose equivalent rate \dot{D}_{max} , only a part of the 896 scenarios would result in a radionuclide inventory that fulfils equation (1). In such a case of dose equivalent rate measurement results more information about the irradiation scenario of the steel block would be required to decide if LSA-I still can be applied or if the block has to be classified differently (e.g. LSA-II).

When the measured dose equivalent rate exceeds \dot{D}_{max} the activity concentration of the steel block exceeds the limit for LSA-I for all scenarios.

It should be kept in mind that in addition also other requirement (i.e. homogeneity of activity for LSA classification) must be respected.

The histogram shown in Figure 2 can be created with RAW-II for all package classifications presented in Table 1. Table 3 presents some practical \dot{D}_{limit} values for a stainless steel cube of $16 \times 16 \times 16 \text{ cm}^3$. It should be emphasized that the stated values are not generally applicable but are based on scenarios found at CERN's high-energy proton acceleratorsⁱ.

Package classification	Exempted material	Excepted material	LSA-I	LSA-II	LSA-III	Type A
D limit at 10 cm [μSv/h]	8.0E-01	1.5E+02	2.0E+01	1.0E+06	2.0E+07	1.5E+05

Tables relating ambient dose equivalent rate limit, and transport class as a function of material type, and dimension have been implemented in the transport logistics tools used at CERN. As a consequence a first pre-classification of goods can be done for a large number of objects in a few minutes. Subsequently, all other ADR requirements such as packaging, documentation etc. can be taken into account in order to carry out the transport according to the ADR.

Conclusions

In order to cope with the large amount of inter-site transports that are seen during shutdown periods at CERN in an ADR compliant manner, an efficient classification method has been developed. Using in-house developed software a large number of nuclide inventories has been calculated for various different material types, geometries and 896 activation scenarios. For each of those configurations the associated dose equivalent rate has been calculated and thus, a relation with the respective transport class have been established. Analysing the large amount of resulting data allowed for establishing envelope scenarios for dose equivalent rate thresholds associated to each transport classification as a function of material type and geometry. Consequently, a fast classification can be done based on dose equivalent rate measurements for a pre-defined set of object classes. After an experimental validation a new organisation has been put in place establishing a new "internal transport office" staffed by two persons. Since November 2018 this "simplified method" has been applied to classify CERN's inter-sites transports using public roads according to ADR.

Within half a year about 5000 goods have undergone this kind of classification and 1000 Class 7 transports have been performed, which clearly shows that such a classification method improves efficiency and can help to optimize radioactive transports considerably.

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