Applied Radiation and Isotopes 122 (2017) 141–147

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

A new approach to characterize very-low-level radioactive waste produced at hadron accelerators

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ARTICLE INFO

Keywords:
Radioactive waste
Characterization
Scaling factor
Correlation factor

ABSTRACT

Radioactive waste is produced as a consequence of preventive and corrective maintenance during the operation of high-energy particle accelerators or associated dismantling campaigns. Their radiological characterization must be performed to ensure an appropriate disposal in the disposal facilities. The radiological characterization of waste includes the establishment of the list of produced radionuclides, called "radionuclide inventory", and the estimation of their activity. The present paper describes the process adopted at CERN to characterize very-low-level radioactive waste with a focus on activated metals. The characterization method consists of measuring and estimating the activity of produced radionuclides either by experimental methods or statistical and numerical approaches. We adapted the so-called Scaling Factor (SF) and Correlation Factor (CF) techniques to the needs of hadron accelerators, and applied them to very-low-level metallic waste produced at CERN. For each type of metal we calculated the radionuclide inventory and identified the radionuclides that most contribute to hazard factors. The methodology proposed is of general validity, can be extended to other activated materials and can be used for the characterization of waste produced in particle accelerators and research centres, where the activation mechanisms are comparable to the ones occurring at CERN.

1. Introduction

The European Organization for Nuclear Research (CERN) operates a number of particle accelerators of high energy. Some beam particles can escape and interact with the accelerators' surroundings and structures, and therefore activate them. At the end of their lifetime, radioactive accelerator components and materials that cannot be reused need to be radiologically characterized prior to their disposal towards a disposal facility. The present study illustrates the process adopted at CERN to radiologically characterize very-low-level (French acronym: TFA – Très faible activité) waste which is eliminated towards the French disposal facility managed by ANDRA (French national radioactive waste management agency).

The chain leading to the final disposal of a batch of waste starts with the identification of the items to be disposed of. Items are included in a batch based on their similarity in terms of activation mechanisms, radiological history and chemical composition. Once a batch is identified and the relevant information collected (i.e. origin of the waste, irradiation history and chemical composition), we calculate the list of the produced radionuclides via either Monte Carlo simulations or analytical codes.

According to reference (IAEA, 2007), we classify each nuclide of the radionuclide inventory as follows:

- Easy-to-Measure (ETM) nuclides, such as γ-emitters, which can be measured via non-destructive nuclear assay means (e.g., in-situ γ-spectrometry)
- Difficult-to-Measure (DTM) nuclides, such as pure-β emitters, which cannot be measured via non-destructive techniques (e.g., they require radiochemical analysis on samples)
- Impossible-to-Measure (ITM) nuclides, such as α-emitters, pure-β and low-energy X-emitters, which are very difficult to measure and which are, therefore, quantified via simulations or calculations.

From the list of ETMs we select the dominant γ-emitter, called Key Nuclide (KN) or tracer. The KN is a nuclide which is systematically measured in each single waste package, with a relative long half-life (years or dozens of years) and whose activity can be correlated to the activity of DTM and ITM radionuclides.

When the radionuclide inventory is available, we collect a repre-

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http://dx.doi.org/10.1016/j.apradiso.2017.01.019
Received 4 November 2016; Received in revised form 20 January 2017; Accepted 20 January 2017
Available online 26 January 2017
0969-8043/

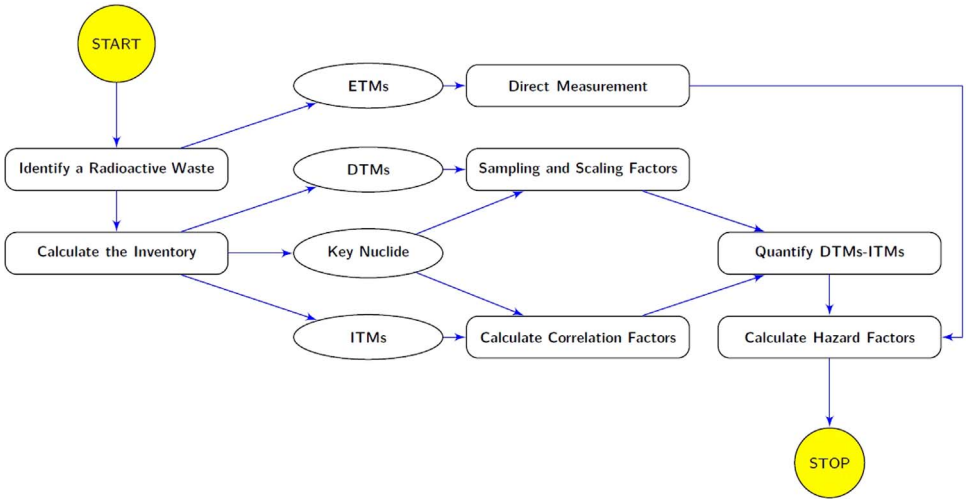

Fig. 1. Radiological characterization process adopted at CERN for very-low-level radioactive waste.

sentative sample to evaluate the activity of the DTMs using the so-called Scaling Factor method (ISO, IAEA, 2009) and perform calculations to estimate the activity of ITMs using the Correlation Factor method (ISO, ISO 16966, 2013.).

We finally measure each single waste package via γ-ray spectrometry and use the activity of the KN to estimate the activity of DTMs and ITMs. The activities so obtained are compared to the limits given by national authorities for waste management to check for the acceptability of the waste in the disposal facilities. Fig. 1 shows a diagram illustrating the principal stages of the process.

Section 2 describes the stages of the radiological characterization process including the methods to quantify ETM, DTM and ITM radionuclides and the waste acceptability criteria. Section 3 presents the results of the calculation of the radionuclide inventories for representative materials activated at CERN together with the application of the characterization method to a metallic waste population. Section 4 discusses the results obtained and the production mechanisms of the main radionuclides produced by activation of metals. Conclusions are given in the last section.

2. Methods

2.1. Identification of waste and calculation of radionuclide inventories

At CERN, a waste elimination campaign starts with the identification of a waste batch. A batch is often chosen based on the volume it occupies in the temporary storage area. When the perimeter of a waste population is defined we start collecting information about its radiological history and chemical composition. Based on this information we calculate the radionuclide inventory using the analytical code Actiwiz (Theis and Vincke, 2012.), which relies on extensive Monte Carlo simulations carried out using Fluka (Battistoni, , 2006; Ferrari, 2005).

When dealing with legacy waste we often have limited knowledge of its radiological history. To predict the complete list of possible radionuclides generated by activation we need to consider a large number of potential scenarios. A scenario is described as the combination of a beam energy, a position inside the accelerator tunnel, an irradiation and a decay time. For the present study we considered beam energies from 160 MeV (Linac 4) up to 7 TeV (Large Hadron Collider), covering the energy spectrum of CERN's proton accelerators. The locations inside the accelerator tunnels span from the beam impact area to the zones behind the concrete shielding. Irradiation and decay times range from a few months to 30 years.

To establish a representative inventory of radionuclides from activated metals we also considered a list of 43 chemical compositions from CERN's materials catalogue (Froeschl and alter, 2012.). We grouped these compounds into 3 families as follows:

- steel, including 16 grades, with density in the range 7–8.75 g/cm^3;
- aluminium, including 7 grades, with density in the range 2.66–2.84 g/cm^3;
- copper, including 20 grades, with density in the range 7.6–8.94 g/cm^3.

For the calculations we used the average value of the concentration of each element, as given by reference standards. The variability of a given element's content is taken into account via the dispersion between the various grades. Table 1 shows a short list of the major elements for common material types used at CERN.

2.2. Quantification of the activity in waste packages

2.2.1. Measurement of easy-to-measure radionuclides

The quantification of ETM radionuclides, including the Key Nuclide, is made via γ-ray spectrometry. This technique is well known and detailed information can be found in numerous classical references (Knoll, 2010; Gilmore, 2008). At CERN, various in-situ and fixed germanium detectors are used for the measurement of γ-emitters in waste packages and samples. The detectors are either nitrogen or electrically cooled and their efficiency for Co-60 at 1.33 MeV spans between 30% and 60%.

We performed a sensitivity study to take into account the variability of various physical parameters such as the apparent density of a waste package, the distance package-detector and the height of the waste within a package. Standard efficiency calibration functions were then created and used to estimate the uncertainty associated with the

Table 1
List of major elements for 4 reference chemical compositions used at CERN. Values in parenthesis are given as weight fractions.

Material grade	Chemical elements and reference concentrations
Aluminium 6060	Al (98.375), Cr (0.05), Cu (0.1), Fe (0.2), Mg (0.475), Mn (0.1), Si (0.45), Ti (0.1), Zn (0.15)
Steel 304L	C (0.03), Cr (18.5), Co (0.1), Fe (67.0825), Mn (2.0), Ni (11.25), P (0.0225), Si (1.0), S (0.015)
Copper OFE	Bi (0.001), Cd (0.0001), Cu (99.99), Pb (0.001), Hg (0.0001), O (0.0005), S (0.0018), Zn (0.0001)
Low carbon content steel	C (0.105), Fe (99.35), Mn (0.45), P (0.02), S (0.025), Si (0.05)