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New methodology for in-situ classification of radiological items with a clearance monitor system

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ABSTRACT

Maintenance activities and operations of high-energy particle accelerators can lead to the collection of radioactive equipment as well as waste materials. In order to ensure their proper classification as radioactive or non-radioactive, one has to quantify the activities of radionuclides produced. According to the regulatory requirements in Switzerland, these activities need to be compared with nuclide-specific clearance limits. In particular, a new set of clearance limits was introduced by the Swiss authorities in January 2018, leading to more conservative values for a number of relevant radionuclides. The present paper complements a previous one in which we developed a methodology to classify equipment with specific characteristics following a dose rate measurement. For equipment that do not fulfill these characteristics, we here extend the methodology by using a total gamma counting device. This methodology concerns the specific material compositions typically found at CERN and takes into account the latest clearance limits introduced by the Swiss authorities. Also, particular considerations are given for electronic components. Their characterization is challenging due to the multiple compositions that can be found in the literature, whereas activation mechanisms are highly sensitive to the material composition.

1. Introduction

Since January 2018, new clearance limits (LL) have been introduced in the “Ordonnance sur la RAdioProtection” (ORAP) [1] and replace the previous exemption values (LE). The ORAP is the Swiss regulation regarding radiation protection. One of the major changes is the fact that LL limits for important radionuclides are often considerably more conservative than LE. For example, the LE limit for Co-60 was 1 Bq/g, whereas the LL limit is 0.1 Bq/g (Table 1). In the case of Mn-54, which is one of the practically most limiting isotopes in metallic components containing iron, the value changed from 10 Bq/g to 0.1 Bq/g.

This paper presents the calculations performed to determine the possibility of classifying materials according to the new LL limits by a total gamma counting measurement and to develop an in-situ radiological classification method [2]. The purpose is to ensure regulatory compliance, keep efficiency in materials classification, optimize and reduce the number of extended measurements.

The classification of potentially radioactive waste has always required a considerable number of measurements at CERN. In the near future, large maintenance campaigns are planned at CERN from 2019–2020 during the so-called LS2 period (Long-Shutdown 2), leading to an increase of the production of waste that is to be classified. Based on the first Long-Shutdown LS1, we extrapolate the number of needed measurements to classify materials as can be seen in Fig. 1.

Table 1
Examples of LE and LL for some radionuclides typically encountered at high energy accelerators.

Isotope	LL [Bq/g]	LE [Bq/g]	Ratio LE/LL
Na-22	1.00E−01	3.00E+00	30
Sc-44	1.00E+01	3.00E+01	3
Ti-44	1.00E−01	2.00E+00	20
Mn-54	1.00E−01	1.00E+01	100
Fe-55	1.00E+03	3.00E+01	0.03
Co-60	1.00E−01	1.00E+00	10

In a previous paper [3], we have shown the possibility to classify potentially activated objects with dimensions above 30 cm x 30 cm x 5 cm with a dose rate measurement. The present paper proposes an extension of the study for objects which do not reach the criteria of [3] and could be non-radioactive. These criteria were identified as:

- Dimensions: ≥30 cm x 30 cm x 5 cm
- Materials: metals except Lead. Concrete and organic compounds are also accepted.
- Equipment containing components with pure Thallium, Lithium or Cadmium shall be excluded (e.g. batteries)
- Mass: ≥10 kg (≥ 4 kg in the case of organic compounds)

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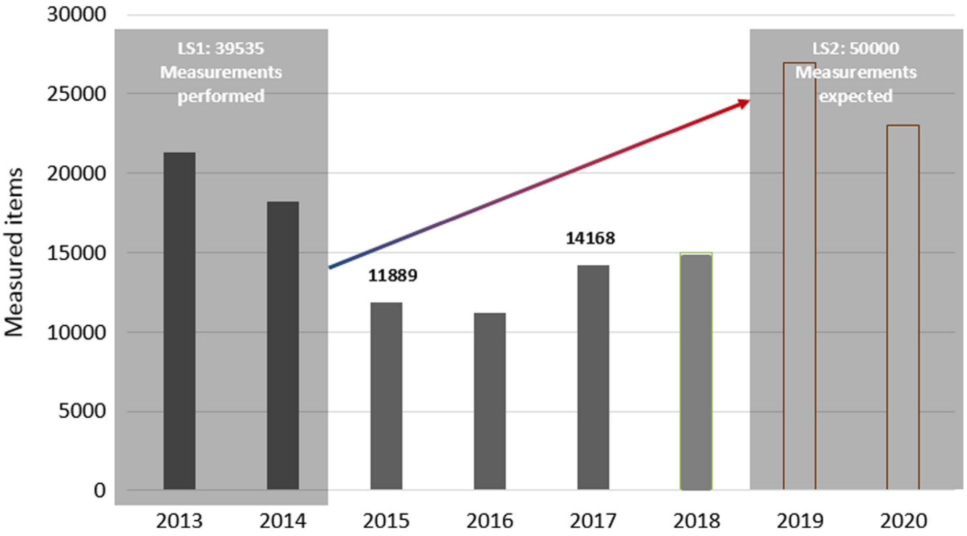

Fig. 1. Expected number of item measurements for LS2 extrapolated from LS1. It should be noted that LS1 lasted for 22 months whereas LS2 will last for slightly more than 2 years. Extrapolation by forecasting.

- Cooling time: ≤2 years (≤4 months in the case of organic compounds)
- Background level: ≤35 c/s with FHZ 512 BGO

Section 2 of this paper details the RADOS device which is used for the materials classification and the calculations performed to construct the so-called radionuclide vectors needed as inputs for RADOS device. Radionuclide vectors are estimates of the most relevant radionuclides produced in a specific scenario, with associated relative concentration. Section 3 summarizes the vectors obtained by calculation. Section 4 treats the mixtures of materials to be classified. Finally, an experimental benchmark is described in Section 5 which is carried out to demonstrate the applicability of the method.

2. Calculation method for RADOS measurements

A similar study than the one for dose rate constraint [3] is done to classify potentially activated objects. The purpose is to use RADOS to measure the activity and deduce whether an object is radioactive or not. In the next, we define generic radionuclide vectors in order to be penalizing. For this purpose, we construct a Figure Of Merit (FOM) that normalizes the toxicity level of a radionuclide vector to the corresponding RADOS detection response.

2.1. RADOS description

RADOS[1] devices are total gamma counting devices used for clearance measurements. Therefore, a nuclide inventory has to be provided to the RADOS software in order to be able to reconstruct the activity values of the radionuclides described in the vector, including alpha or beta emitters that are not directly detectable. RADOS devices are produced by Mirion Technologies. One of the specimens in use at CERN is shown in Fig. 2.

The device contains large area plastic scintillation detectors. A lead shielding ensures low radiation background inside the measurement chamber, with a thickness between 3 cm for the door and 5 cm on sides, top and bottom. The object mass is determined during the measurement process to convert detected activity into mass specific activity. Further detailed information about the RADOS device can be found in [2].

Fig. 2. RADOS RTM 661/440 lnc at CERN.

2.2. ActiWiz calculations

In order to compute a Figure Of Merit (FOM — see Eq. (3)) to identify the most penalizing scenario and consequently, the most penalizing radionuclide vector for each material, an extensive set of calculations is done with ActiWiz 3.3.15/2018 – 1010 [4].[2]

The considered scenarios are the following:

- All locations in the accelerator, from the beam line to behind the shielding,
- All machines with primary beam energies from 160 MeV to 7 TeV as well as the four major LHC experiments (ATLAS, LHCb, CMS, ALICE) [5],
- Irradiation times from 1 day to 30 years,
- Cooling times from 3 h to 2 years.[3]

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[1] https://mirion.s3.amazonaws.com/cms4_mirion/files/pdf/spec-sheets/rtm661-440-premium-class-clearance-monitor.pdf?1523977410.

[2] http://actiwiz.web.cern.ch/.

[3] We note in passing that the present study is not applicable for historic waste which requires specific calculations. It rather focuses on a generic approach for materials that have cooling times that are at maximum similar to the length of long shutdown periods like LS2 etc.

- Material composition for the most typical objects, *i.e.*, aluminum (cf "Aluminum_6060"), iron ("Iron_ARMCO"), stainless steel ("Steel_304L"), copper ("Copper_CuOFE"), Inconel ("Inconel-0.21-Co"), concrete ("Concrete_CERF_EU") and polyethylene ("Polyethylen"). The elemental compositions are provided in the CERN ActiWiz catalog [6,7]. In addition, particular attention is given to printed circuit boards considering two different material compositions identified in the literature, based on electronics scrap which was mixed, shredded and analyzed to get an average composition. These compositions are identified as "Electronics_Kindesjo" [8] and "Electronics_Goosey" [9]. The characterization of this highly specific equipment has been tested and extensively verified with experimental data in a specific study [10].

The present study comprises 80 000 calculations of nuclide inventories and the corresponding radiotoxicity expressed in multiples of LL. From these quantities, the FOM (see Eq. (3)) is constructed as follows:

The summation rule value (S_LL, see Eq. (1)) is being calculated to obtain the respective radiotoxicity in terms of LL fraction. It is the sum of the specific activity a_i divided by the radiotoxicity level LL_i for all the radionuclides i in presence:

$$S\_LL = \sum_i \frac{a_i}{LL_i} \tag{1}$$

where a_i is the specific activity per primary of the radionuclide i, $\mathrm{LL_i}$ is the clearance limit regarding the radionuclide i [1].

Then, we calculate the RADOS response (Eq. (2)) as:

$$Response = \sum_i a_i * lnc_i \tag{2}$$

Where

$\mathrm{lnc_i}$ is the Leading Nuclide Correlation (LNC) factor of the radionuclide i as defined and detailed in [2]. RADOS devices are calibrated with a Co-60 source and detected net count rate is converted into Co-60 activities. LNC factors are developed by RADOS and correlate the detector response in Co-60 equivalent activity to the activity of a given radionuclide.

The FOM is assessed dividing the summation rule (S_LL) of a scenario by the RADOS response for this scenario as follows (see Eq. (3)):

$$FOM = \frac{S\_LL}{Response}\left[\frac{g}{Bq}\right] \tag{3}$$

Eventually, for each material the scenario giving the higher FOM corresponds to the most penalizing one, as the S_LL is maximized for a minimized RADOS response.

The results and associated scenarios for each material are summarized in Table 2. The most penalizing case in terms of material is, similar to the finding in Ref. [3], Aluminum. Due to the close FOM regarding the iron-based materials (Iron, Inconel and Stainless Steel) it is decided to group these materials to limit the number of nuclide vectors. For the same reason, printed circuit boards (PCB) are grouped together (Electronics_Kindesjo [8] and Electronics_Goosey [9]). Hence, we consider the most penalizing scenario for the classification of the three iron-based materials as well as the most penalizing scenario of the two electronics compositions (see Section 3).

For polyethylene, the RADOS response is driven by the Be-7. However, due to the short half-life of this radionuclide (53 days), the RADOS response is very sensitive to the respective cooling time. For this reason, the penalizing cases found for polyethylene are split in different cooling time scenarios ranging from 2 months to 1 year in order to evaluate the feasibility of the measurement at different stages of the decay of Be-7.

The RADOS MDA (Minimum Detectable Activity) is typically below or equal to 50 Bq equivalent Co-60 [2] with the RADOS RTM 661/440 lnc. Depending on the mass of the object measured, the MDA will not be sufficient to reach the limit (S_LL) and then the RADOS may give a false positive result (considering an object as radioactive whereas it is not). For the sake of completeness, we evaluate with Eq. (4) the minimum mass needed to ensure that the MDA is sufficient to reach the limit (S_LL).[4]

$$M_{min} = FOM\left[\frac{g}{Bq}\right] * (MDA + \underbrace{1.96\sqrt{MDA}}_{uncertainty\ at\ 1.96\sigma})[Bq] \tag{4}$$

For instance, with the MDA and FOM given above, the minimum mass of aluminum needed to be able to establish that an object is not radioactive with the RADOS RTM 661/440 lnc would be 2.26 kg whereas for copper the minimum mass would be 1.2 kg. Below this minimum mass, the RADOS gives the result that the object is radioactive and, if the actual mass is very well below the minimum mass, the RADOS software gives a warning stating that the mass is too low to reach the limit.

3. Radionuclide vectors proposed

Based on the most penalizing scenario for each material (see Table 2), we construct the most penalizing radionuclide vector with the following constraints:

- Contribution to S_LL is above 99%,
- Contribution to RADOS response is above 99%.

Each vector is given with a total activity normalized to 100%.

3.1. Aluminum

The radionuclide vector for Aluminum is presented in Table 3. The main contributor to activity is Zn-65 which is also the main contributor to the RADOS' response (88%) and the S_LL value (97%). It should be noted that Na-22 can also be identified as a significant contributor to RADOS' response (10%).

Table 2

Summary of the most penalizing scenario for each material. Electronic materials (Electronics-Goosey and Electronics-Kindesjo) as well as iron materials (Iron_ARMCO, Inconel-0.21-Co and Steel 304L) have been grouped in the subsequent analysis, retaining the most conservative composition with the higher FOM.

Mat	Energy	Position	Irradiation	Cooling	FOM
Aluminum_6060	ALICE	outside-L3	1mo	6mo	35.2
Electronics-Goosey	ATLAS	TAGGER	1d	1.5y	23.4
Iron_ARMCO	160 MeV	10cmTarget	1y	2y	22.1
Inconel-0.21-Co	160 MeV	BeamImpact	1y	2y	21.5
Electronics-Kindesjo	160 MeV	WithinBulky	2m	1.5y	21.0
Steel_304L	160 MeV	BeamImpact	1y	2y	20.8
Concrete_CERF_EU	ALICE	outside-L3	30y	2y	19.0
Copper_CuOFE	160 MeV	BeamImpact	1d	2y	18.7
Polyethylen	160 MeV	CloseWallBeamOnBulky	30y	<= 2m	5.8
				<= 6m	15.2
				<= 1y	126.7

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[4] MDA+1.96SQRT(MDA) is an approximation to evaluate the confidence interval which is calculated slightly differently in the RADOS system (i.e., according to ISO11929).

Table 3
Normalized radionuclide vector for Aluminum. The fraction of the activity inferred from the measured signal is given for each radionuclide of interest.

Na-22	Cr-51	Fe-59	Zn-65
0.024	0.055	0.005	0.916

3.2. Iron based compounds

The radionuclide vector for iron-based compounds is presented in Table 4. For this specific case (we retain IRON_ARMCO as the most penalizing material (see Table 2)) Mn-54 is the main contributor to S_LL and the RADOS' response with more than 99%.

Table 4
Normalized radionuclide vector for iron-based compounds. The fraction of the activity inferred from the measured signal is given for each radionuclide of interest.

Mn-54
1

3.3. Copper

The radionuclide vector for Copper is presented in Table 5. The radiotoxicity in terms of S_LL is driven mainly by Mn-54 (40%), Co-60 (24%), Co-57 (18%) and Zn-65 (15%). Regarding the RADOS' response, the main radionuclides are Co-60 (45%) and Mn-54 (35%).

Table 5
Normalized radionuclide vector for Copper. The fraction of the activity inferred from the measured signal is given for each radionuclide of interest.

Mn-54	Co-56	Co-57	Co-58	Co-60	Zn-65
0.151	0.008	0.689	0.006	0.092	0.055

3.4. Concrete

The radionuclide vector for Concrete is presented in Table 6. The europium radionuclides represent together 98% of the contribution to the RADOS' response and 99% of the S_LL value.

Table 6
Normalized radionuclide vector for Concrete. The fraction of the activity inferred from the measured signal is given for each radionuclide of interest.

Na-22	Eu-152	Eu-154
0.006	0.777	0.217

3.5. Polyethylene

The radionuclide vectors for Polyethylene are presented in Table 7. After 6 months of radioactive decay, H-3 is the main contributor to the radiotoxicity in terms of S_LL (74%), whereas Be-7 is the only contributor to the RADOS response. With Be-7 having a relatively short half-life (53 days), the radionuclide vector of polyethylene is split in three sub-vectors, depending on the cooling time, from 2 months to 1 year.

Table 7
Normalized radionuclide vector for Polyethylene. The fraction of the activity inferred from the measured signal is given for each radionuclide of interest.

Cooling	H-3	Be-7	C-14
2mo	0.8767	0.1228	0.0004
6mo	0.9711	0.0284	0.0005
1y	0.9967	0.0028	0.0005

3.6. Electronic components

In [10], we study an average composition based on Goosey's composition (see [9]) and show that ActiWiz can be used to do the predictions. Some differences were found but are explained in [10]. We show that, for Ag-110m, Mn-54, Co-57, Co-60 and Zn-65, the gamma spectrometry measurements performed at CERN on electronic components from 2004 to 2019 agree with the ActiWiz calculations quantitatively. We found discrepancies for Co-58, Be-7, Cr-51 and Na-22. For these latter radionuclides, we here study the associated FOM, presented in Table 8. Those radionuclides can be traced back to a bias coming from short cooling times due to measurements done shortly after irradiation whereas the calculations consider equal distribution between scenarios with long and short cooling times. For more details, the reader is invited to refer to [10]. We see that the whole set of radionuclides has a FOM below the one of the most penalizing electronic scenario (FOM = 23.4, see Table 2). Hence, the discrepancies observed in [10] cannot negatively affect the classification results (i.e. considering as not radioactive an equipment that is radioactive).

Table 8
Radionuclides for which activity distributions differs between ActiWiz and gamma spectrometry distributions.

Radionuclide	LL (Bq/g)	RADOS response (Bq/g)	FOM
Co-58	1	0.54	1.85
Be-7	10	0.03	3.33
Cr-51	100	0.02	0.50
Na-22	0.1	1.06	9.43

Moreover, for Na-22, the RADOS response remains high (1.06 Bq/g) then this radionuclide would still be detected by the RADOS and its LL is of 0.1, which is the same as Zn-65/Ag-110m/Mn-54 (representing 95% of the activity). Consequently, a wrong estimation of Na-22 activity in electronics components would lead to spread its activity (detected by RADOS) to radionuclides with same LL and then would not affect the classification process.

However, discrepancies observed on Na-22 between measurements and calculations in [10] come from electronic components that are inside an aluminum container. In the case an electronic component contains aluminum, the production of Zn-65 would increase. With a FOM of 39, an increase of this radionuclide in the electronic composition would lead to an increase of the FOM of the electronic component and consequently a penalization of the radionuclide vector. Hence, we suggest to consider the aluminum nuclide vector described for the RADOS measurement process instead of electronics vector when electronics components with aluminum equipment are classified.

Also, a list of 10 radionuclides is not predicted when considering Goosey's composition with ActiWiz whereas they are found in gamma spectrometry results [10]. The reason is some missing element in the electronics composition. These radionuclides are summarized in Table 9 with their associated LL and RADOS response.

Table 9
Radionuclides found in gamma spectrometry not appearing in ActiWiz calculations.

Radionuclide	LL (Bq/g)	RADOS response (Bq/g)	FOM
Se-75	1	0.17	5.88
Rb-83	1	0.29	3.45
Y-88	0.1	0.92	10.87
Te-123m/Sc-47	1/100	0.041/0.034	24.39/0.29
Ta-182	0.1	0.52	19.23
Sc-46	0.1	0.97	10.3
Sc-47	100	0.034	0.3
Br-82	1	1.36	0.7
Sb-122	10	0.26	0.4
Sb-124	1	0.77	1.3

For all these radionuclides, except Te-123m, the associated FOM is below the one of the most penalizing electronic activation scenario (FOM = 23.4, see Table 2). For this reason, the introduction of these radionuclides in the composition cannot lead to a more penalizing result. For the sake of completeness, we analyze the impact that the 83 most common trace elements with radiological relevance would have on the FOM when mixed with Goosey's composition. This mixture process is performed in ActiWiz using the same process as described in next Section 4, with mass concentrations from 1E-03% to 10% of the potentially problematic element. We observe a decrease of the FOM of all the scenarios when we mixture electronics compositions with elements identified in [10] and described in Table 9 which are leading to the creation of these radionuclides. Consequently, they would have no effect on the classification and can be neglected.

As a consequence, the Goosey composition given in [9] can be used for all electronic components that are not mainly made of aluminum or not contained in an aluminum box.

The radionuclide vector for electronics is presented in Table 10. The radionuclide vector contains six radionuclides. The S_LL is dominated by Zn-65 with 81% of contribution, then Ag-110m with 13% of contribution. Regarding the RADOS response, the main contributors are Zn-65 (49%) and Ag-110m (42%).

Table 10
Normalized radionuclide vector for electronics. The fraction of the activity inferred from the measured signal is given for each radionuclide of interest.

Ag-110m	Mn-54	Co-57	Zn-65	Co-60	Na-22
0.124	0.044	0.037	0.782	0.008	0.004

4. Mixture of materials

Metallic objects can be made of one single type of metal that might contain impurities, or they can be constituted by an assembly of different types of metal, each with their own impurities. The case of an assembly of different types of metals is fully covered by the case of an object made of aluminum, which is more conservative than any of the other metals considered. Nevertheless, the study is extended to cover 85 possible chemical compositions by systematically adding stable chemical elements as impurities (the 85 elements supported by ActiWiz), leading to over 565'000 activation scenarios.

The study aims at quantifying the problematic elements in case of encountering a mixture by weight of 99%, 95%, 90% or 70% Aluminum_6060 with respectively 1%, 5%, 10% or 30% of 85 chemical elements available in ActiWiz.

The FOM for the mixture is computed as follows (Eq. (5)) for a specific activation scenario:

$$FOM_{mixture} = \frac{wt * S\_LL_{element} + (1 - wt) * S\_LL_{aluminum\_6060}}{wt * Response_{element} + (1 - wt) * Response_{aluminum\_6060}} \tag{5}$$

Where

S_LL$_{\text{element}}$ and S_LL$_{\text{aluminum\_6060}}$ are the summation rule for LL (Eq. (1)) for a specific scenario regarding respectively the activation of the studied element and the aluminum_6060 material;

Response$_{\text{element}}$ and Response$_{\text{aluminum\_6060}}$ the RADOS response respectively for an activated element and for the aluminum_6060 material in a specific scenario (Eq. (2));

wt is the mass fraction of the element in the mixture.

The results with the chemical elements causing an increase of FOM when mixed with Aluminum are presented in Tables 11 and 12 below.

Only 20 elements over the 83 could be penalizing for the classification as the FOM increases when they are added to the Aluminum composition.

Gases, such as Nitrogen and Xenon (Table 11) should be excluded by the procedure as well as heavy metals such as Bismuth, Gadolinium, Thallium, Thulium, Thorium, Tungsten and Uranium as these categories shall anyway be treated separately due to their densities. These elements will be treated with a different methodology that could involve dedicated Monte Carlo simulations and gamma spectroscopy analyses.

Table 11
Penalizing chemical elements when mixed with Aluminum and associated FOM variations. Elements to be excluded from the measurement process.

Radionuclide	Weight fractions				
	0%	1%	5%	10%	30%
Lithium	35.19	217.62	1040.5	2171.95	8314.13
Thallium	35.19	191.52	904.21	1883.36	7182.87
Samarium	35.19	95.14	394.87	788.30	2608.64
Cadmium	35.19	68.64	262.00	527.00	1898.00
Thulium	35.19	59.88	136.89	198.03	361.63
238-U	35.19	35.26	57.04	94.84	284.56
Nitrogen	35.19	35.27	49.28	79.38	242.78
235-U	35.19	61.24	62.84	63.24	63.57
Xenon	35.19	35.19	45.19	50.82	56.41
Bismuth	35.19	35.69	37.81	40.73	56.24
Gadolinium	35.19	35.69	37.77	40.60	55.48

Table 12
Penalizing chemical elements when mixed with Aluminum and associated FOM variations. Elements to be considered in case of mixture of materials.

Radionuclide	Weight fractions				
	0%	1%	5%	10%	30%
Chlorine	35.19	35.45	36.58	38.14	51.59
Barium	35.19	35.28	35.67	40.96	47.6
Tungsten	35.19	35.28	35.62	36.11	38.63
232-Th	35.19	33.52	33.09	33.03	38.54
Zinc	35.19	38.05	38.42	38.47	38.51
Calcium	35.19	35.21	35.28	35.37	35.87
Erbium	35.19	35.20	35.23	35.27	35.50
Sulfur	35.19	35.19	35.22	35.24	35.38
Helium	35.19	35.2	35.21	35.24	35.36

Table 13
Normalized radionuclide vector proposed for a mixture by weight of 70% aluminum and 30% chlorine. The fraction of the activity inferred from the measured signal is given for each radionuclide of interest.

Na-22	Cr-51	Fe-59	Zn-65	S-35
0.000037	0.0019	0.000041	0.0017	0.9963

Due to their high FOM when introduced, even at 1%, Lithium, Cadmium and Samarium have to be explicitly excluded by the procedure (Table 11). Indeed, including them in the radionuclide vector for mixture of materials would lead to a very penalizing vector that does not allow classifying any material. Hence, objects containing these elements have to be removed from the measurement process and other solutions need to be identified for the classification, such as Monte Carlo simulations and gamma spectroscopy analyses.

Among the remaining penalizing chemical elements (Table 12), the maximum FOM is 51.6 for Chlorine with a 30% mixture in Aluminum. The associated RADOS vector for such a mixture with Aluminum_6060 at 70% in mass and Chlorine at 30% in mass is described in Table 13. This vector is suggested to be used as the vector for material containing unknown elements at a maximum of 30% in mass, under the condition that heavy metal, gases and Lithium/Cadmium/Samarium exclusions previously listed are respected. Materials containing these elements should not be part of the classification process.

For this specific nuclide vector the RADOS' response is dominated by Zn-65 (82%), Na-22 (6.8%) and Cr-51 (6.5%). The radionuclides which are the main contributors to the radiotoxicity in terms of S_LL are Zn-65 (61%) and S-35 (37%).

5. Validation with an experimental benchmark

In order to validate the method an accompanying experiment is carried out. The objective is to irradiate objects (Aluminum and Iron slabs of 2 kg) and reach a radiotoxicity value of S_LL = 1. Subsequently, the proposed RADOS measurement-based classification is carried out and the conclusion verified with gamma-spectrometry.

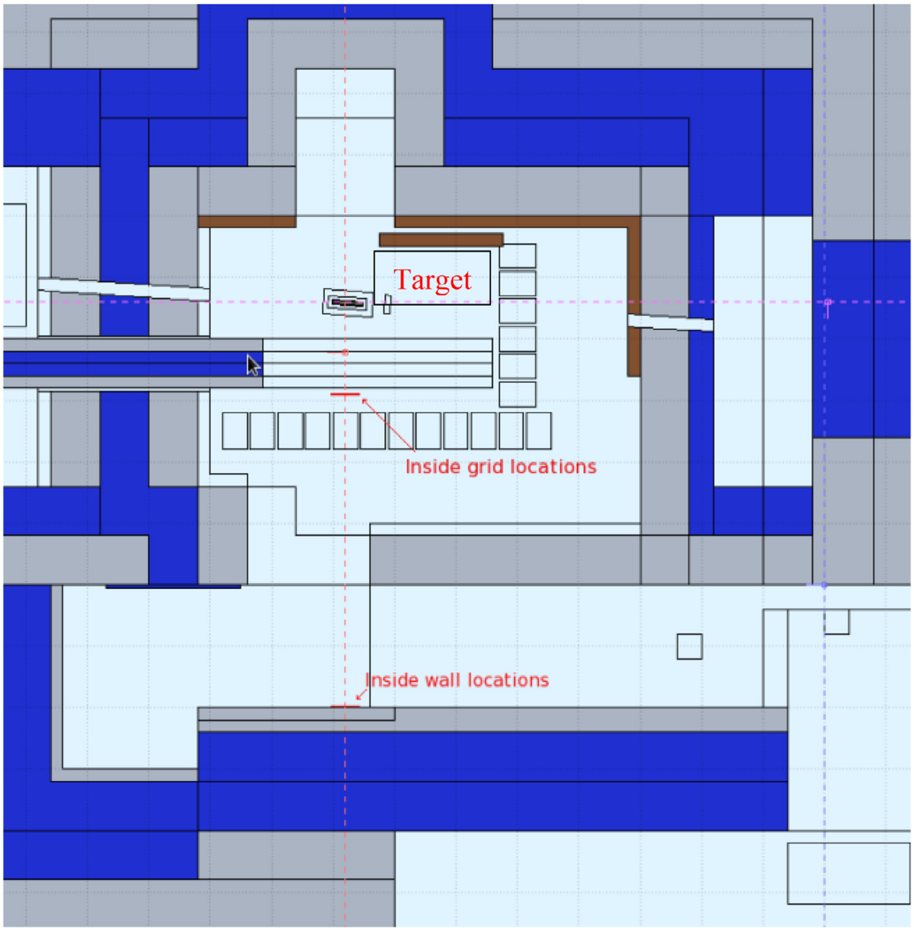

Fig. 3. Section of the FLUKA geometry of the CHARM facility, including also an indication of the irradiation locations. The irradiated objects are located at the position labeled "inside wall locations".

5.1. Experimental setup

The irradiation of the material is performed in CERN's CHARM [11, 12] facility in the East Experimental Area of the Proton Synchrotron (PS).

In CHARM a beam of protons with a momentum of 24 GeV/c is impacting on a target, leading to the creation of secondary particles. The characteristics of the proton beam are well known and the number of protons impinging on the target is recorded in a database.

The two slabs are positioned adjacent to the target where the proton beam impacts (Fig. 3 — "inside wall locations" and Fig. 4).

The fluence of secondary particles at the location of irradiation is simulated with FLUKA [13,14]. These data in combination with the beam intensity are subsequently used in ActiWiz Creator [15] to calculate the respective nuclide inventories. As a consequence, a prediction of the irradiation and cooling time required to reach a radiotoxicity level of S_LL = 1 is carried out for each material type.

5.2. Experimental data

Irradiation patterns are planned based on ActiWiz calculations carried out to retain activation levels that remain sufficiently low and allow for reaching a RADOS response above the MDA (usually 50 Bq equivalent Co-60 with the RADOS RTM 661/440 lnc used).

Spectrometry measurements are performed to verify specific activities and the associated radiotoxicity levels (S_LL). In Table 14 below the comparison of the calculated and the measured results for the two samples is given.

Fig. 4. Samples of aluminum and iron irradiated in CHARM.

In general, the values predicted by the ActiWiz calculation show good agreement with the gamma-spectrometry measurements, in particular for the main contributors to the RADOS response and S_LL.

Observed discrepancies remain below a factor of 4 yielding very good agreement of the RADOS response values obtained by measurement (RADOS, gamma-spectrometry) and calculation.

Table 14
Comparison of calculations/measurements of specific activities of samples after their irradiation in CHARM. Only radionuclides which contribute to more than 99% of RADOS response or 99% of the S_LL are presented. S_LL and RADOS response are calculated with Eq. (1) and Eq. (2). The RADOS specific activities are calculated with the vectors for Iron based compounds and Aluminum, presented respectively in Sections 3.2 and 3.1.

2 weeks after irradiation							
28/09/2018				10/01/2018			
IRON				Aluminum			
RN	γ-spectrometry (Bq/g)	ActiWiz (Bq/g)	RADOS (Bq/g)	RN	γ-spectrometry (Bq/g)	ActiWiz (Bq/g)	RADOS (Bq/g)
Sc-44	6.07E−03	2.02E−03	–	Be-7	3.98E−02	1.40E−01	–
Sc-46	3.50E−03	4.18E−03	–	Na-22	5.69E−02	5.61E−02	1.65E−02
V-48	4.51E−02	6.39E−02	–	Sc-46	3.38E−03	4.28E−03	–
Cr-51	1.57E−01	2.10E−01	–	V-48	8.06E−03	1.64E−02	–
Mn-52	5.71E−02	7.33E−02	–	Cr-51	4.15E−01	9.66E−01	3.78E−02
Mn-54	6.92E−02	7.46E−02	4.68E−01	Mn-52	7.79E−03	7.72E−03	–
Co-56	1.84E−03	3.94E−03	–	Mn-54	2.23E−02	2.69E−02	–
Fe-59	2.08E−02	9.00E−02	–	Zn-65	1.72E−02	7.63E−03	6.30E−01
Eq.Act.Co60	0.20	0.30	0.21	Eq.Act.Co60	0.12	0.14	0.18
S_LL	0.87	1.06	4.68	S_LL	1.05	1.00	6.46

We see that RADOS vector contributes to maximize activity for some radionuclides, *i.e.* Mn-54 for Iron and Zn-65 for Aluminum, which are the most penalizing radionuclides of the samples.

The comparison shows that the S_LL evaluation is penalizing with the RADOS vectors presented in Section 3, as planned, with a factor of 5 in average in the present experimental cases studied.

6. Conclusion

Following the introduction of new LL limits, the classification of potentially activated materials at the exit from the accelerators can be performed on the basis of a total gamma counting measurement, by using a RADOS RTM detector [2].

In this paper, we present a method to conservatively be able to judge about the radiotoxicity level (S_LL) of CERN's equipment with a RADOS device. This method is based on the assessment of the Figure Of Merit (FOM, i.e. the S_LL level per RADOS response) of a wide range of plausible scenario at CERN. Based on the most penalizing scenario identified for each material type, we construct a set of penalizing radionuclide vectors to classify metallic, concrete, electronics or burnable activated materials.

The method is here applied to CERN using tools such as FLUKA and ActiWiz. ActiWiz is developed specifically to CERN particle's fluence spectra. However, the method described in this paper can easily be used for other installations worldwide, considering other tools for the calculations, such as CINDER [16], FISPACT-II [17], GEANT4 [18], MCNPX [19]).

In case of mixture of materials, the most penalizing vector shall be used and, in addition, a very conservative vector is eventually proposed for material containing up to 30% of unknown chemical elements.

The case of electronic components is studied thanks to chemical compositions found in the literature [8,9] and a validation of these compositions [10]. Such components containing aluminum should be measured in RADOS device considering the aluminum radionuclide vector.

These vectors can be used for objects which respect some acceptance criteria:

- Absence of cadmium, lithium, samarium, glass (not studied), permanent magnets, batteries, gas or heavy metals (bismuth, lead, tungsten, thallium, thorium, thulium and uranium);
- The cooling time shall not exceed 2 years and specific vectors based on the cooling times shall be used for burnable materials.

A validation study is performed with two metallic slabs irradiated at CERN in the CHARM facility and the comparison between the calculations, the RADOS measurements and the gamma spectrometry analysis shows consistent results and demonstrates that the vector used with the RADOS are penalizing as expected by the methodology of this study.

Objects which fail to meet the above acceptance criteria would need complementary measurements or additional calculations in order to be correctly classified.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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