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Full Length Article

Radiological clearance of historical waste from particle accelerators

Lucie Svihrova [a,b,*], Kerstin Bauer [a], Luca Bruno [a], Gerald Dumont [a], Matteo Magistris [a], Nabil Menaa [a], Marco Silari [a], Luisa Ulrici [a]

[a] CERN, Geneva 23, 1211, Switzerland
[b] Faculty of Nuclear Sciences and Physical Engineering, Czech Technical University, Prague, 11519, Czech Republic

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ABSTRACT

This paper presents a detailed methodology for reclassifying radioactive material from particle accelerators as non-radioactive, drawing on experiences at CERN. During particle accelerator decommissioning, the waste is classified based on its radioactivity level where the very-low-level activity material is the potential clearance candidate. Complying with the Swiss legislation, the waste must fulfil three criteria in order to be cleared. Before performing the radiological measurements, the waste must be processed (disassembled, cut, …) due to its size and multi-material composition. To properly evaluate the measurement results, a detailed theoretical study must be performed providing information on e.g. expected radionuclides. As a part of quality assurance, the theoretical models are verified using gamma spectrometry measurements on waste samples. The presented methodology is supported by the summary of past and ongoing CERN projects as well as know-how learned by years of experience.

1. Introduction

The life cycle of a particle accelerator includes several phases: design, construction, commissioning, operation, decommissioning, dismantling, and disposal, where the latter may occur many years after the end of operation. The radioactivity induced in the accelerator and in its ancillary components during operation can be minimized by an appropriate choice of materials at the design stage (e.g., avoiding as far as feasible the use of steels with comparatively high cobalt content), but decommissioning inevitably involves the disposal of activated – or potentially activated – materials. These may come not only from the machine but also from the surrounding infrastructures and shielding, and are classified as radioactive material. The materials are typically solid, but sometimes there are liquids as well (e.g., oil from vacuum pumps).

The disposal implies assessing even very low levels of radioactivity in the individual parts of the accelerator and its ancillary equipment. This subject has grown in importance in recent years, and still little is found in the literature on radioactivity predictions in view of accelerator decommissioning and disposal of material. Apart from the group's previous work [1], only specific parts of radiological clearance methodology are addressed in available publications: assessing of residual radioactivity from 600 MeV synchro-cyclotron at CERN [2], overarching concerns of decommissioning, such as radiological assessments and the economic impacts, crucial for facilities like CERN [3], verifying compliance with regulatory standards through specific methodologies including gamma spectroscopy [4], insight into the activation processes and waste classification at a granular level, applicable to similar accelerators at CERN [5], and treatment methods, particularly electrochemical techniques, enhancing the waste management process were studied [6]. Additionally, the costs and radiological implications of dismantling EU accelerators is evaluated [7], focusing on waste volume, decommissioning expenses, and strategies for minimizing activation

Most of the radioactive material that is generated in particle accelerators can be classified as material with very-low-level (VLL) [8] activity, which can also be a candidate for clearance from regulatory control (also called free-release). Notable exceptions would be targets, beam-dumps and any other accelerator components that are directly hit by the beam and therefore can reach low- to intermediate-levels of activity. The disposal of VLL activity waste towards final repositories requires accurate radiological characterization to ensure that the activity they store falls below the activity limits for which the repository was designed. The waste characterization relies on a range of measurement techniques, but also on calculations that require information on the irradiation history of the waste. The radiological characterization of historical waste - and therefore its successful elimination towards a final repository - is particularly challenging because the information on its irradiation history may have been partially lost over the years.

* Corresponding author at: CERN, Geneva 23, 1211, Switzerland.
E-mail address: lucie.svihrova@cern.ch (L. Svihrova).

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The possible disposal pathways depend on the national regulations. In Europe, the European Commission (EC) provides directives [9] to be adopted by its member states. Each country is allowed to introduce further regulations and laws as compared to the EC requirements. For instance, until February 2022 [10,11], the French legislation did not provide clearance limits below which waste coming from a radiological area can be treated as non-radioactive and cleared from the regulatory control. Release of such waste was only allowed after a detailed theoretical study supported by extensive experimental measurements to produce a detailed "zoning" of the accelerator and experimental areas, i.e., a classification of areas where material could or could not have been activated. This was the approach adopted at CERN at the time of the Large Electron–Positron Collider (LEP) decommissioning in 2000 [12]. The application of clearance is complementary to the zoning approach, still effective at CERN, for material and waste irradiated in the zone of activation. The International Atomic Energy Agency (IAEA) [13,14] also provides standards and guidelines for clearance and release of waste.

The IAEA Nuclear Energy series technical report on "Decommissioning of Particle Accelerators" [15] summarizes the decommissioning strategies, organization and technologies for particle accelerators and provides extensive information on waste management and specifically on clearance of waste from the regulatory framework. The example of the LEP decommissioning is reported in its annex as one of the national examples for accelerators of class IV (energies in the range of GeV to TeV).

This paper describes guidelines to follow for the radiological characterization of VLL activity material, based on the experience obtained at CERN. The process includes:

- Recovery of available background information, such as mass and elemental composition of the various materials, the sources of activation, the irradiation history, etc.;
- Extensive gamma spectrometry measurements on samples of all materials, complemented by radiochemical, ambient dose equivalent rate ($\dot{H}^*(10)$) and contamination measurements, to anticipate the expected levels of the residual radioactivity;
- Monte Carlo simulations (e.g., with the FLUKA code [16,17]) and analytical calculations (using ActiWiz [18]) to supplement the experimental results and to determine the list of expected radionuclides — radionuclide inventory. The Monte Carlo study gives an insight into the activation processes, allowing the application e.g. of the "authoritative sampling technique", where the samples are chosen based on a higher probability of being activated
- An operational phase during which systematic radiological measurements of all items are performed. Such measurement results must then satisfy the criteria enabling the clearance of the waste from regulatory control. In Switzerland, three criteria are defined in the Radiation Protection Ordinance (RPO) [19]:
  1. The ambient dose equivalent rate, $\dot{H}^*(10)$, at 10 cm from the surface must be below $0.1\ \mu Sv/h$ after subtraction of natural background.
  2. If there is a possibility of person contamination, the surface contamination c_i of a given radionuclide i must be below a surface contamination guidance value CS_i defined in the RPO. If a mixture of radionuclides is present, the surface contamination summation rule given by expression (1) must be satisfied:
  $$\sum_i \frac{c_i}{CS_i} < 1. \tag{1}$$
  3. The specific activity A_i of a given radionuclide i must be below a clearance limit LL_i (Limite de libération) or the total activity must be lower than the activity of 1 kg of a waste whose specific activity is equal to the clearance limit. As well as for the surface contamination, if a mixture of radionuclides is present, the summation rule given by expression (2) must be satisfied:
  $$\sum_i \frac{A_i}{LL_i} < 1. \tag{2}$$
  Naturally Occurring Radioactive Material (NORM) radionuclides are not considered in the calculation of the summation rule but are individually compared to the relevant NORM clearance limits LLN [19].

2. Clearance projects at CERN

Prior to evaluating the suitability of waste for a project of clearance from regulatory control, CERN systematically evaluates two major aspects:

- Candidate volumes: the expected volume of waste that is candidate for clearance might be relatively small (i.e. a few m^3). In this case, the cost required to demonstrate that clearance requirements are met can be disproportionate, and it can even exceed disposal costs as radioactive waste. In this case, it is advisable to wait for a future clearance campaign, and include such small amount of waste in a larger batch.
- Availability of information: if there are large uncertainties on the radiological history of the waste (which might originate by an intentional delay in the waste processing for elimination or by errors), conservative assumptions need to be made. These assumptions can be very penalizing, and they can potentially lead to non-radioactive material being classified as radioactive.
- Sufficiency of resources: it is necessary to ensure the availability of adequate personnel with the requisite skills, alongside the provision of suitable workplaces, and radiological measurement capabilities.

For more than 10 years CERN has undertaken a series of projects aiming to systematically characterize historical waste for its final disposal. A first example is the partial decommissioning in 2010 of the 600 MeV synchro-cyclotron, the first CERN accelerator in operation from 1957 to 1990. After 20 years of radioactive decay, the facility underwent a partial decommissioning: the accelerator hall was converted into a Visit Point with the machine on display, but more than 200 tons of waste were radiologically characterized and eliminated as either VLL activity waste or free released [2]. Since 2016, CERN has undertaken further campaigns to clear from the regulatory control some of its historical waste, such as accelerator equipment set aside after decommissioning to let it decay for several years before its final disposal, possibly as non-radioactive waste in Switzerland. These projects are briefly summarized here below:

- Project CLEAR (**C**haracterization of **LE**P **A**cceleration **R**F system) was the first project of this campaign. The elimination of the superconducting radio-frequency acceleration system of the former LEP collider, has been described in an earlier paper [1].
- Project ELISA (**ELI**mination of **S**hredded c**A**bles) was focused on the elimination of 40 tons of shredded Oxygen Free Electric (OFE) copper from power and control cables, which were used in supervised and controlled radiation areas and subsequently removed in the period between 1982 and 2014. About 75% of the material could be released as non-radioactive immediately, the rest was divided into three groups requiring 5, 10 and 15 years of further radioactive decay to bring the residual activity below the (present) clearance limits of $0.1\ Bq/g$ for ^{60}Co and $100\ Bq/g$ for ^{63}Ni.
- Project CLELIA (**C**learance of the **LE**P **L**ead-**I**nsert **A**luminium chambers) proceeded to treat old LEP equipment, namely about 150 tons of an aluminium/lead mixture from the vacuum system plus about 30 tons of steel, galvanized steel and tungsten.

- Project AMELIA (**A** LEP **M**agnet **ELI**mination **A**ctivity) treated a number of LEP magnets (24 dipoles, 98 quadrupoles and 21 sextupoles) for a total mass just above 600 tons.
- Project PLATAN (**PLA**stic **TAN**ks from evaporated CNGS water) eliminated 5.3 tons of plastic tanks previously filled with infiltration water from the CERN Neutrinos to Grand Sasso facility (CNGS). This project is currently extended to a larger variety of type and origin of the water.
- Project AMAL (**A**utre **MA**teriaux **L**EP; in French: Other LEP Materials), recently completed, is the clearance process of about 106 tons of mixed LEP equipment, a miscellaneous of vacuum pumps, HV resistors, magnets, small RF cavities, collimators, single-metal components, etc.
- Project CRANES (**CRANES**), currently ongoing, treats about 81 tons of material from few overhead cranes formerly installed in the Intersecting Storage Rings (ISR) collider and in the ECN3 hall in the North Experimental area served by the Super Proton Synchrotron (SPS).

Overall, about 95% of the old LEP equipment could be cleared as non-radioactive across all projects. The residual 5% of waste was classified as VLL activity waste, but mostly because of the conservative measures taken during the clearance process (for example, by lowering the Swiss $\dot{H}^*(10)$ limit for clearance from $0.1\,\mu\mathrm{Sv/h}$ above background at $10\,\mathrm{cm}$ down to $30\,\mathrm{nSv/h}$ above background at contact, see Section 5).

All clearance projects, executed at CERN so far, created a net economical gain or were cost neutral. The cost–benefit analysis was determined on the one hand by the mass of recycled valuable materials, and on the other hand by the avoided expensive disposal of radioactive waste volumes. Among the most important costs in the process of clearance, typically, about 75%–90% of the clearance budget is allocated to manpower (characterization studies, radiological measurements, expert technical support, waste handling, project management, etc.), the rest covering tools, equipment, consumables, and elimination of a small amount of radioactive waste.

3. Characterization study

For any new project an individual study of the equipment history and induced activity is performed to confirm its feasibility. The results are a necessary input for radiological characterization.

3.1. Induced radioactivity in accelerators

Residual radioactivity represents the main source of exposure of personnel working at high energy particle accelerators, as exposure to prompt radiation is reduced to practically negligible levels by the shielding, and prohibited access to the beam areas by means of interlocks during operation. Induced radioactivity depends on many factors: the type and energy of the accelerated particles, the beam intensity as well as the composition of the materials irradiated by the primary beam and the secondary radiations.

Distributed radioactivity levels at proton accelerators are considerably higher than at electron accelerators [20]. Induced radioactivity at proton accelerators is due to high-energy nuclear reactions induced by neutrons and protons: intra-nuclear cascade generating spallation products, fragmentation reactions, π-meson production, high-energy fission, etc. [21,22].

At electron accelerators the reactions involved include spallation reactions, photonuclear reactions by intermediate energy photons and low-energy neutron capture [23,24]. Short-lived radionuclides (with half-lives ranging from a few minutes up to several hours) are only of concern for maintenance interventions during the operational phase, where the individual exposures can be strongly reduced or avoided by simply sufficiently delaying the intervention. Medium- and long-lived radionuclides (half-lives ranging from a few days to several years) are the main source of exposure for delayed interventions, and only the longest-lived ones are of concern at the time of decommissioning and for the final disposal of the infrastructure, when the environmental impact becomes the dominant aspect to be coped with.

3.2. Theoretical characterization

A theoretical characterization study is necessary to determine the radionuclide inventory of the material, and to establish a strategy and define the parameters for radiological measurements. Only then, the specific activity can be determined and used as the third criterion to decide about clearance.

In order to group items into batches with similar radiological properties, their former use is researched. Information about their origin, location and irradiation time in the accelerator is of fundamental importance in this respect. Representative items, randomly selected, are radiologically measured and weighted, their material composition is determined either theoretically (in case of availability of proper documentation) or verified by X-ray fluorescence measurements. Their precise elemental composition is then taken from the list of materials used at CERN [18]. Materials present in negligible amounts that do not contribute to the radionuclide inventory can be then excluded from the study.

Pre-calculated irradiation scenarios can be directly selected in the ActiWiz tool [18] for current CERN high-energy proton accelerators and for several representative locations. For all other cases, a detailed evaluation of the irradiation is first performed using the Monte Carlo simulations package FLUKA [16,17], to calculate the spectral fluence of the secondary particles. The geometry is simplified and historically known energies and particle types are used as the primary beam. Several scoring volumes are chosen in the item to obtain a full picture of the activity distribution. The secondary particle spectra are then used as an input in the ActiWiz Creator software [18]. After that in both cases, other parameters are selected, including elemental composition, irradiation, cooling times and the purpose of the calculation, which in our case outputs a list of radionuclides ordered by their contribution to the summation rule.

As the complete list of nuclides that can be created in a material can be extensive, in some countries, e.g., Switzerland, the radionuclides that need to be considered in the radionuclide inventory for the declassification of material can be limited to those that contribute to 90% of the summation rule [19].

In case of combined irradiations raising from use of the material in several facilities, all situations are independently studied and the strictest one (resulting in the highest summation rule) is considered. Ion pumps from the AMAL project are one such example, since before their use in LEP some of them were employed in the ISR. It was shown that the ISR irradiation is negligible compared to the one in LEP.

From the radiological characterization point of view, the radionuclides that make up the radionuclide inventory can be classified as Easy- and Difficult-To-Measure (ETM and DTM, respectively). ETM radionuclides are typically gamma emitters, which can be measured by non-destructive techniques, such as gamma spectrometry. The evaluation of the activity of DTM radionuclides can be performed experimentally, but it requires destructive techniques and radiochemical analysis. Alternatively, the activity of a DTM nuclide can be inferred from the activity of one ETM nuclide (also called the Key Nuclide, KN) if a correlation between the activities of the DTM and of the KN exists. In this case, the activity of the DTM nuclide can be calculated as the activity of the KN, multiplied by a so-called Scaling Factor (SF) [25].

The SF method was initially developed for the management of radioactive waste produced by the operation of nuclear power plants. The required correlation between DTM and KN can only be established if the radiological history and the production mechanisms are the same for the waste considered. In case of nuclear power plants, the consistent production of nuclides with correlated activities is the consequence of

Table 1
Commonly expected ETM and DTM radionuclides for materials typical for the projects. For each scenario the list of radionuclides must be determined separately due to different irradiation/cooling parameters as well as material impurities.

Material	ETM radionuclides	DTM radionuclides
Aluminium	^{22}Na	
Cables	$^{44}Ti/^{44g}Sc$, ^{60}Co, ^{125}Sb	
Copper	^{60}Co, ^{108m}Ag	
Stainless steel	$^{44}Ti/^{44g}Sc$, ^{60}Co	
Steel	$^{44}Ti/^{44g}Sc$, ^{60}Co	^{3}H
Titanium	$^{44}Ti/^{44g}Sc$	

the steady operation of the reactors, which share similar materials and operating conditions.

The application of the SF method to historical radioactive material from particle accelerators is at the same time challenging and innovative. High-energy particle accelerators are typically unique prototypes which utilize a wide range of different technologies and materials — as opposed to nuclear reactors of the same type. In addition, the radiological history of produced waste is often unknown, and cooling times after the end of irradiation can span from a few years to over 30 years. Because of this variability, it is not possible to guarantee that – in the general case – a correlation exists between DTM and KN nuclides within a given batch of radioactive material. Such correlation can only be predicted by systematic calculations, which need to cover hundreds of possible activation scenarios, and it requires experimental confirmation by extensive sampling.

The list of radionuclides expected in an item of waste, with their contribution expressed as a fraction of the total activity, is called fingerprint. Table 1 gives examples of commonly expected radionuclides for different materials.

A Figure of Merit (FOM) [26] is introduced to compare different activation scenarios and to select the one which would lead to the most penalizing classification in case of measurement with the total gamma counter (see Section 5):

$$\mathrm{FOM} = \frac{\sum_i \frac{A_i}{LL_i}}{\sum_i A_i \cdot LNC_i}, \quad (3)$$

where A_i is the specific activity and LL_i the corresponding clearance limit of radionuclide i. The LNC_i ("Leading Nuclide Correlation") [26, 27] is a parameter reflecting the gamma detection efficiency of energies emitted by the radionuclide i and is provided by the manufacturer of the total gamma counter. The nuclide vector with the highest FOM is the most conservative and is thus chosen for the calculation.

3.3. Experimental characterization

At the beginning of a project, the items are grouped into batches either by type (e.g., vacuum chambers, ion pumps), their irradiation history, or by their main material composition. Hazardous materials (liquids, chemicals, asbestos, etc.) and components requiring a specific study other than the one covered by the project (such as batteries or printed circuit boards) are removed from the project as out of the scope.

In parallel to the theoretical characterization (see previous Section 3.2), experimental measurements are performed. $\dot{H}^*(10)$ and contamination screening measurements allow identifying the materials that fulfil the key requirements for being candidates for clearance, as well as planning the subsequent operational procedure to minimize the radiological hazard for the personnel that will be handling the items. Moreover, several random samples are taken and analysed by gamma spectrometry. For verification purposes, the list of detected radionuclides is compared to the calculated radionuclide inventory.

The activity of DTM radionuclides can be experimentally determined by radiochemical analysis performed at laboratories outside CERN. As an example, for the ELISA project, more than hundred samples were sent to AMEC Foster Wheeler, United Kingdom, to quantify besides other radionuclides the ^{63}Ni activity. The results confirmed that the ActiWiz calculations were conservative because most of the experimental SF for ^{63}Ni and its KN ^{60}Co were well below the calculated values.

4. Treatment techniques in view of characterization

Following the characterization study, the equipment which was stored at CERN may need to undergo manual processing before the radiological measurements. The decision on whether or not to disassemble the equipment, and up to which level, depends on the material classification and on the cost/benefit factor.

Some of the clearance projects treated large and complex pieces of equipment, such as the superconducting radiofrequency acceleration system in the CLEAR project [1] or large dipole, quadrupole and sextupole magnets from the AMELIA project. This equipment was systematically disassembled by trained personnel following a well-defined procedure. Separated pieces were grouped together according to the material type and placed in containers suitable for the radiological analyses. Based on the experience at CERN, the manual dismantling cost for clearance is estimated to be equivalent to the manpower cost required by the conditioning of the material for elimination as radioactive waste. A different approach was employed for the ongoing project CRANES, where some of the large pieces were cut using a hydraulic press in order to fit into containers.

As an opposite case, the AMAL project mostly covered smaller pieces of equipment for which individual studies were needed. Supports and waveguides are the examples of material whose disassembling essentially only required removing several screws, after which the material was separated, measured and sold individually. Equipment like bellows or RF tuners, which are small and light and whose material composition is well known, were treated without disassembly. Equipment such as ion pumps or collimators stand between these two extremes: they were partially disassembled by removing ancillary components, but a full disassembly would either be technically difficult or would not bring any benefit, neither as financial profit nor as advantage for the characterization.

Another treatment technique that can be utilized for clearance activities is shredding. A cable shredder enabling to shred the cable and to separate the metal (typically copper) from the insulation was used for the cables treated in the ELISA project. The insulation and the copper were studied separately. The copper shreds were homogenized, stored in drums and sampled to determine the residual radioactivity. However, the characterization of insulation alone is complicated because it offers virtually no ETM radionuclide that could be used as KN for the application of the SF method. For future projects, it was therefore decided to characterize whole cables without shredding or separation of insulation from metallic wire. Indeed, the cost of shredding would not be compensated by an adequate increase in the resale price.

In the case of permanent magnets from the AMAL ion pumps, an additional treatment step was included because their magnetic field may affect the functionality of the handheld measurement devices as well as the total gamma counter. The 8.1 tons of magnets, which could potentially be sold once cleared, therefore underwent a thermal treatment in order to demagnetize them. They were loaded in several batches into a large oven and heated to at least 900 °C, which is higher than their Curie temperature. After the treatment, no remaining magnetic field was detected and they could follow the usual measurement procedure, after which 96% of them were classified as clearable.

Melting of activated material with a goal to lower the specific activity was also tested by CERN. In 2005 a pilot project of processing 14 tons of material was undertaken, followed by a full-scale project of 270 tons in 2007. The process of revalorization of VLL activity metallic material involved its melting in a specific melting installation for radioactive scrap, STUDSVIK Radwaste, Sweden (nowadays Cyclife Sweden AB) and its recycling after radiological checks. The aim of the project was the proof of concept for the homogenization of the residual activity within the waste material matrix and the removal of impurities. In fact, some of the impurities that include radionuclides responsible for the residual activity of the material can remain in the slag, to be treated as radioactive waste at the end of the process. By means of clearance, the project of melting reduced the amount of radioactive material to be disposed of from 120 m^3 to 1.8 m^3. The technique is effective for contaminated metallic waste, however nowadays it brings associated cost and environmental load, so it can be often replaced by alternative clearance techniques.

It should be mentioned that any of these treatment techniques are never meant to mix or dilute the activated mass with additional non-active material, a practice forbidden in the Swiss legislation unless the explicit approval of the licensing authority is provided for the material up to 10 times the LL. The aim is to either segregate the fraction of the material that shows a residual activity exceeding the clearance limits, or to make the matrix of the material more homogeneous, in order to ensure that each and every part of the cleared waste satisfies the requirements set by the regulatory guidelines.

In case a chemical risk is also present, extra precautions must be taken. The risk of lead contamination was present during the CLELIA project, during which aluminium vacuum chambers surrounded by a few millimetres of lead shielding were handled. For the equipment manipulation, a well confined lead risk area was created, that could only be accessed by properly trained personnel equipped with the required personal protective equipment. A similar approach was adopted during the AMAL project, when asbestos discs attached to stainless steel components were systematically removed and sent to a specialized CERN laboratory for processing. If the presence of asbestos is suspected, the material is isolated from the rest and a chemical expert is asked for assistance. The radiological clearance of radioactive material containing asbestos is planned as a future project.

At CERN the same types of tools, including complex machinery, are used on material eligible for clearance and on low- to intermediate level activated waste with no contamination. Whenever cleaning is technically feasible and economically affordable, a unique processing tool (e.g., hydraulic press-shears) is used for both clearance projects and for the disposal of ascertained radioactive waste. This process ensures that no dust, created by the processing of the radioactive material, would stick to the equipment undergoing radiological clearance.

5. Radiological measurements and analysis

After the preliminary classification and sorting of material, each item undergoes a set of radiological measurements and analyses, starting with $\dot{H}^*(10)$ and surface contamination measurements. Then the items are grouped, stored into standard containers, and their specific activity is analysed by total gamma counting. Afterwards, quality control analyses are performed and if they are successful, all measurements are documented in a protocol to be submitted to the Swiss safety authorities. The process of the radiological measurements and analyses is depicted in Fig. 1.

5.1. $\dot{H}^*(10)$ measurements

For the $\dot{H}^*(10)$ measurements, a sturdy calibrated device with a sensitivity down to a few tens of nSv/h must be used. At CERN the instrument typically employed is the Automess AD6/H+AD b/H [28], calibrated by the Swiss accredited Institut de Radiophysique (IRA) in Lausanne. For places difficult to access, such as the inside of vacuum chambers, additional measurements with a more compact device are performed. A BGO scintillation probe (Thermo FHZ 512 [29]) attached to a digital survey meter (Thermo FH40GL [30]), calibrated at CERN, serves the purpose. Both devices, BGO and AD6/H+AD-b/H are calibrated using a ^{137}Cs source.

At the beginning of each measurement period, the natural $\dot{H}^*(10)$ background is measured, which should not exceed 80 nSv/h as its small error enhances the certainty in measuring our target threshold. Each item is measured individually or in batches, depending on its size, either by slowly scanning its surface or measuring at predetermined points. While choosing the measurement points, parameters such as self shielding effect, measurement duration, and locations more likely to be activated are considered.

Although the $\dot{H}^*(10)$ clearance limit set by the Swiss authorities is 100 nSv/h at 10 cm distance, a stricter value of 30 nSv/h at contact is used for radiological clearance at CERN. This stricter threshold allows the majority of VLL activity material to be identified already during the first measurement. This approach also prevents the risk of triggering false alarms at the truck portal monitor, where all material is analysed before exiting CERN, due to the so-called mass effect.

5.2. Contamination measurements

As second type of radiological measurement, each item undergoes direct measurement for both alpha and beta/gamma contamination. A CoMo 170 [31] contamination monitor (calibrated at IRA for the expected radionuclides) is used at CERN. Even though alpha contamination is not expected in waste undergoing radiological clearance, it is routinely checked using ^{241}Am as reference radionuclide. For beta/gamma contamination, ^{60}Co is chosen as a reference radionuclide. The contamination of each item is directly measured at the same locations previously chosen for the $\dot{H}^*(10)$ measurements or by a scan at contact with the surface, and subsequently compared to the thresholds. We used to adapt the guidance values given in the Swiss legislation [19], however, recently we adopted stricter thresholds provided by the Agreement concerning the international carriage of dangerous goods by road (ADR), valid in 53 countries including EU and Switzerland [32], specifically 0.4 Bq/cm^2 for beta/gamma contamination and 0.04 Bq/cm^2 for alpha contamination. For all ongoing and future projects we ensure that the stricter regulation are being observed.

After these measurements, the items are sorted by type or by material composition and stored in wire-mesh containers of approximately 0.7 m^3 volume; the mass of each container's content should not exceed 100 kg (see Section 5.3).

In addition to the direct contamination measurement which can be affected by the signal coming from inside the material, an indirect measurement is performed by a smear test sample taken from an area of 100 cm^2. The alpha and beta activity transferred to the smear sample is analysed by an LB4200 Multi-Detector Low Background Alpha/Beta Counting System [33] and corrected by the smeared area and standardized 10% transfer factor [34] to obtain the surface activity of the original smeared item. Two smear test samples are taken for each grid container. In case of difficult-to-access locations, an additional smear test sample is taken. For the indirect contamination analysis results, the same thresholds are applied as for the direct contamination measurement [32].

For some types of materials, such as multi-layer insulation, static electricity present on the measured material can affect the direct contamination measurement results by giving incorrect higher values. In such cases, only the indirect contamination analysis results are considered.

5.3. Specific activity measurements

The third criterion for releasing the items, based on their specific activity, must also be fulfilled. At CERN a total gamma counter is used whenever possible. The clearance monitor RTM644lnc™ from Mirion Technologies [35] is designed for reliable clearance measurement of material and equipment of various geometries and sizes, ranging from small to large objects such as pallets, waste bags, grid boxes, 200 l

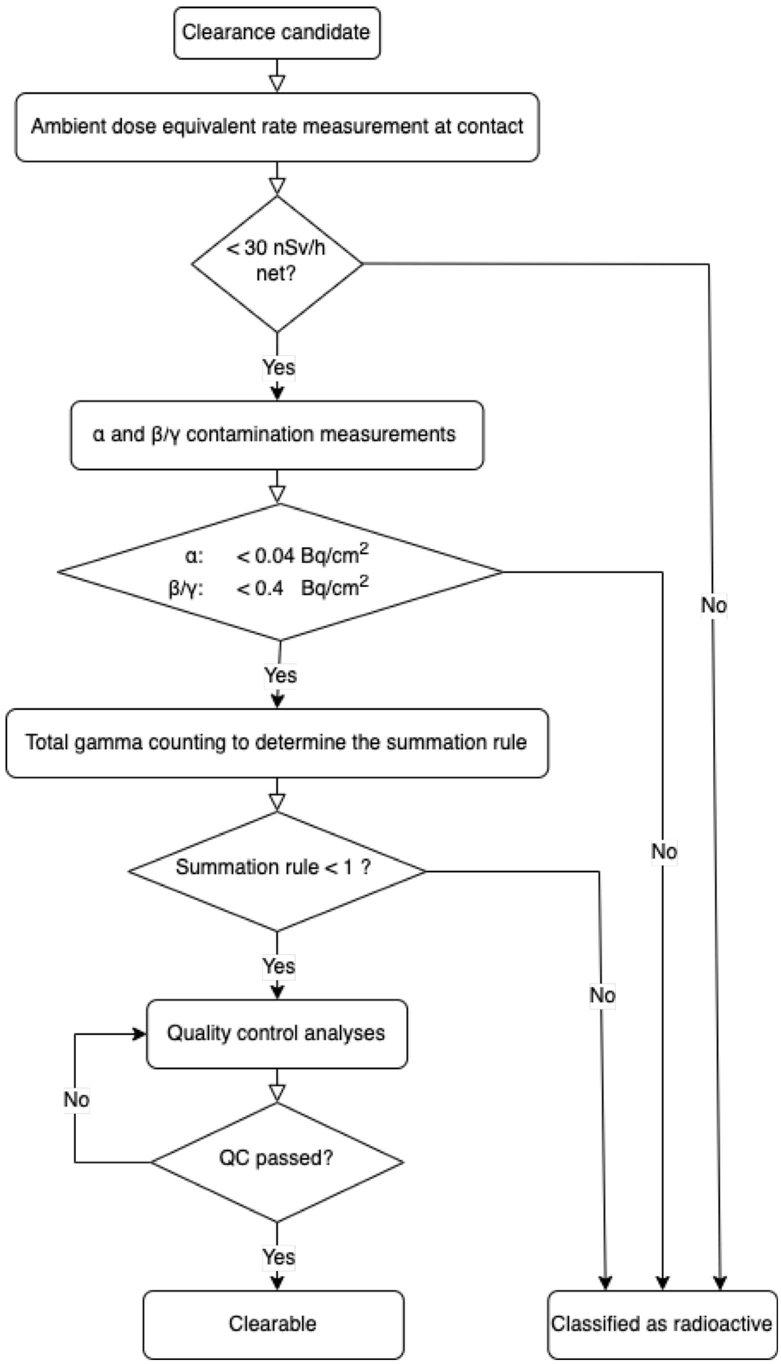

Fig. 1. Radiological characterization flowchart.

and 400 l drums. The integrated 24 large-area detectors offer an approximately 4π geometry, ensuring high sensitivity, low detection limits and recognition of significant inhomogeneities. The monitor is shown in Fig. 2.

At the beginning of each set of analyses, a "closed chamber" measurement is performed to evaluate the ambient background. Afterwards, each container is individually measured and analysed by the clearance monitor. The software converts the total gamma counts into ^{60}Co equivalent activities, considering calibration factors that include both mass and geometry of the items. One of the geometry parameters is the filling level, which in combination with the mass allows to estimate the items' density and thus the self-shielding effect. For the clearance measurements, the filling level is often set to 1%–25% – the most conservative choice – because then the calibration corresponds to the highest density and thus the highest self shielding effect. The ^{60}Co equivalent activities are then converted into radionuclide activities based on the given fingerprint (see Section 3) using the LNC factors provided by the manufacturer. The specific activity is obtained by dividing the total activity by the net mass of the container content. If the net mass exceeds 100 kg, the specific activity is obtained conservatively by dividing the total activity by 100 kg to fulfil the Swiss legislation criteria of the maximum averaging mass [19]. The specific activities are then used for the calculation of the summation rule (2), the third criterion is satisfied when the result is lower than one. The clearance monitor gamma counting analysis is made more conservative by intentionally including counts of the NORM radionuclides, which can be present in the material but do not have to be considered in the summation rule [19].

Fig. 2. Large clearance monitor RTM644lnc™.

5.4. Quality assurance

5.4.1. In-toto gamma spectrometry of grid boxes and large items

To verify the radionuclide inventory, the 10% containers having the highest summation rule, as determined by the clearance monitor analyses, are analysed using High Purity Germanium (HPGe) detectors Falcon 5000 [36] from Mirion technologies (see Fig. 3). Each detector undergoes regular on-site verification and quality assurance to ensure continuous quality and reliability during the detector operation. The gamma spectrometry analyses are performed in a dedicated facility of the radiological analysis laboratory. In Situ Counting Object System (ISOCS) and Laboratory Sourceless Calibration Software (LabSOCS) from Mirion Technologies (Canberra) [37,38] are used for efficiency calibration of various waste geometries as shown in Fig. 4.

In case any of the detected radionuclides are not in the radionuclide inventory, their presence is investigated, and the clearance activity is suspended until the characterization study is re-evaluated and, if needed, updated.

The summation rule can also be calculated from the gamma spectrometry results and exceptionally, upon approval from the Swiss authorities, it can substitute the clearance monitor analyses if it cannot be performed e.g., for geometry reasons.

5.4.2. Laboratory gamma spectrometry analyses of samples

At least one sample is randomly selected from each container to be analysed by gamma spectrometry. Typical samples are screws, bolts, and nuts, as well as easily cuttable parts like cables or pipes. When several containers have the same content, different materials and samples are taken to achieve sample variability. The samples undergo gamma spectrometry measurements and analyses at CERN's radio-analytical laboratory, which is equipped with several HPGe detectors from Mirion Technologies, mainly SEGe GC4018 P-Type [39]. The detector's relative efficiency [40] at 1332 keV ranges from 40 to 60% and the Full Width at Half Maximum (FWHM) is approximately 1.2 keV at 122 keV and around 2 keV at 1.33 MeV. The sample mass, geometry and gamma spectrometry geometries and acquisition times are defined to guarantee acceptable MDA [41] values with respect to the clearance limit.

5.5. Clearance decision and organization

At the end of the clearance process of each batch of material, the material is stored in a so-called "buffer zone" in the Radioactive Waste Treatment Centre. Once the protocol with the results of all radiological measurements has been approved by the Swiss authorities, the material is repackaged into transport containers and moved to a dedicated area to be picked up by a scrap dealer.

5.6. Truck global monitoring

The last measurement is performed just before leaving the CERN site, when each truck passes through one of the sensitive Thermo Scientific FHT 1388 S Modular Radiation Portal Monitors [42], situated on the two main CERN sites, Meyrin and Prévessin.

This additional measurement provides the assurance that the total activity of the material is below the local radiation background. In particular the mass effect (very low residual activity on several packages can add up to become detectable above the background) is tested. The measurement done at CERN, previous to the material leaving the site, lowers the chance that the content triggers an alarm at the scrap dealer's premises and is returned to CERN. It also constitutes as a reference if additional radioactive material is added to the content of the truck during the transport by a third party.

6. Conclusions

This paper gives an overview of the methods and tools applied at CERN for the radiological clearance of VLL activity material produced in the decommissioning of particle accelerators. We have focused our attention on high-energy accelerators, but these methods and tools are equally applicable for lower-energy machines, for example the growing number of cyclotrons and synchrotrons employed for particle therapy with protons and carbon ions.

For a given clearance campaign, the selection of candidate items is performed according to the following criteria:

- the type and level of residual radioactivity expected (radionuclide inventory),
- practical considerations (storage optimization, treatment techniques needed for the characterization and sorting of the material, chemical toxicity, the availability of reliable information on the irradiation history, etc.), and
- the resources (budget, equipment, etc.).

If the material is eligible for clearance from regulatory control, i.e., to be disposed of as non-radioactive, a theoretical characterization study is needed to demonstrate that the material is at least likely to fulfil the requirements in terms of radionuclide inventory, limits for specific activities, and thresholds for $\dot{H}^*(10)$ and contamination values. The manual processing and experimental radiological characterization then require detailed measurements on all material: gamma spectrometry analyses on representative samples, total gamma counting on individual components or batches of components, $\dot{H}^*(10)$ as well as direct and indirect contamination measurements. A final check is performed with a sensitive truck portal monitor before the material leaves the site. This measurement also serves as an independent verification because scrap dealers are normally equipped with a truck portal monitor to check incoming scrap.

In terms of labour and budget, clearance of scrap metal is typically cheaper than elimination as weakly radioactive waste towards national repositories. Elimination of radioactive waste, irrespective of

Fig. 3. Gamma spectrometry setup in a dedicated area.

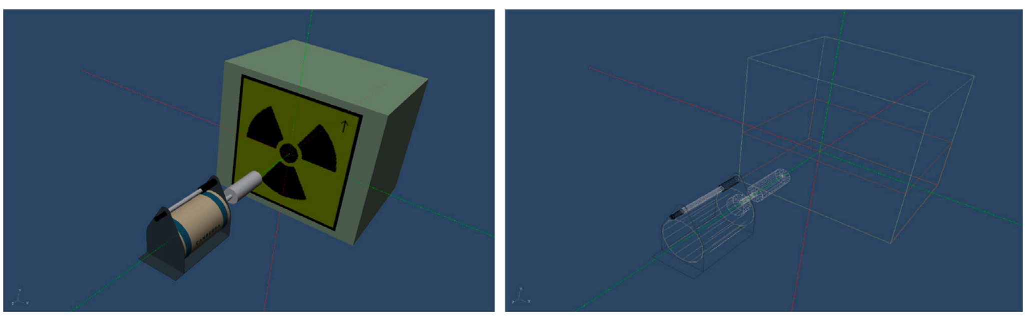

Fig. 4. ISOCS 3D model rendering of the reference geometry.

Table 2
Amount of rock m_{rock} that would have to be excavated to extract the same mass of metals m_{metals} recycled by CERN radiological clearance projects CLEAR, ELISA, CLELIA, AMELIA, and AMAL from 2016 to July 2022 using ore grade [43,44] - the share of ore that is useable metal. For iron, an estimation that it makes up 100% of steel components and 70% of the stainless steel components was made.

Element	Ore grade [%]	m_{metals} [tons]	m_{rock} [tons]
Aluminium	19.0	261	1 636
Copper	0.9	104	11 615
Iron	40.0	804	2 815
Lead	60.0	72	191
Nickel	0.5	7	1 407
TOTAL		1 238	16 415

how weakly radioactive it is, requires substantial costs for the packaging work, the dedicated containers, the transport, and the final storage. The sale of scrap metals also brings some revenues, which can partially or fully cover the elimination costs of the clearance project.

The most important aspect to be considered, especially nowadays, is perhaps the environmental footprint: recycling metals spare the work to extract ores from mines and process them, eliminating the associated risks and the impact on the environment. For CERN clearance projects to date, this impact can be estimated using data on the percentage of ore that can be used as metal in the case of aluminium, copper, iron, lead and nickel [43,44]. Table 2 compares the quantity of material released with the mass of rock that would have to be excavated to provide an equivalent amount of pure element if these metals would have not been revalorized.

CRediT authorship contribution statement

Lucie Svihrova: Writing – review & editing, Writing – original draft, Software, Methodology, Investigation, Formal analysis, Data curation. **Kerstin Bauer:** Writing – review & editing, Writing – original draft, Software, Methodology, Investigation, Formal analysis, Data curation. **Luca Bruno:** Writing – review & editing, Writing – original draft, Methodology. **Gerald Dumont:** Writing – review & editing, Validation, Supervision, Methodology, Investigation. **Matteo Magistris:** Writing – review & editing, Writing – original draft, Validation, Methodology, Formal analysis. **Nabil Menaa:** Writing – review & editing, Writing – original draft, Software, Methodology, Formal analysis. **Marco Silari:** Writing – review & editing, Writing – original draft, Validation, Supervision, Resources, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization. **Luisa Ulrici:** Writing – review & editing, Supervision, Methodology, Investigation, Conceptualization.

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.

Data availability

Data will be made available on request.

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