

Radiological characterization of high-energy proton Tantalum targets from CERN-ISOLDE Facility

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Abstract—Accelerator-based techniques are considered among the leading methods to produce radioactive nuclei. The ISOLDE facility (Isotope mass Separator On-Line DEvice) at the European Organization for Nuclear Research (CERN) is a unique source of beams of radioactive nuclides that are used in a wide range of research domains, from nuclear astrophysics to life sciences. Over 400 ISOLDE targets are currently stored at CERN and an average of 30 targets are irradiated every year. All these targets are planned to be dismantled and disposed of as radioactive waste in a dedicated repository in Switzerland. This paper provides an overview of the challenges related to the radionuclide activity predictions, the number of activation scenarios, dismantling and conditioning of the Radioactive Waste (RW) using a hot cell (HC), the high dose rate, uncertainties related to unknown geometry parameters of the RW packages, and finally industrializing a complex RW elimination process as ITEP for an accelerator complex. The performed work addresses each of these challenges and offers technical solutions based on state-of-the-art computational codes, statistical techniques, and state of art Non-Destructive Assay (NDA) techniques using high energy-resolution gamma spectrometry. The methodology followed can be of guidance for the development of similar processes at other facilities.

the MEDICIS project was initiated by CERN in 2010 [3][4]. The CERN-MEDICIS experimental facility (MEDical Isotopes Collected from ISOLDE) is dedicated to the production of non-conventional radionuclides for research and development in medical imaging, diagnosis, and radiation therapy. Most of the MEDICIS operation is driven by the ISOLDE facility from which it receives protons with the same energy and intensity.

Over 300 ISOLDE targets are currently stored at CERN and an average of 30 targets are irradiated every year. All these targets are planned to be dismantled and disposed of as radioactive waste in a dedicated repository in Switzerland following the tripartite agreement between CERN, France and Switzerland (Host States) [5]. The first elimination phase consists of dismantling 12 so-called Tantalum targets and categorize them into structural (Aluminum), target core (Tantalum), and other structural materials. The resulting materials are packaged in dedicated waste drums depending on the material type and undergo a complete characterization. The corresponding radiological acceptance criteria require an accurate knowledge of the levels of induced radioactivity and contamination of the targets. The radiological characterization of the ISOLDE and MEDICIS targets is therefore a necessary step in view of their disposal.

II. MATERIALS AND METHODS

The following sections describe the ISOLDE and CERN-MEDICIS targets, hot cell (HC) dismantling and drum packaging, and methodologies used to estimate and quantify the radionuclide inventory.

A. ISLODE and CERN-MEDICIS Targets

At the ISOLDE facility, radioactive ion beams (RIBs) are produced via spallation, fission, or fragmentation reactions in a thick target, irradiated with a proton beam from the Proton Synchrotron Booster (PS-Booster) at an energy of 1.4 GeV. What is usually referred to simply as “target” is a complex assembly that comprises the target core and its container, the ion source, the target base, and the auxiliary components. Since each physics experiment requires a different target and ion source configuration, different target units are manufactured and irradiated every year. Figure 1 shows a 3D drawing and a

I. INTRODUCTION

THE ISOLDE facility (Isotope mass Separator On-Line DEvice) at CERN (European Organization for Nuclear Research) is a unique source of beams of radioactive nuclides that are used in a wide range of research domains, from nuclear astrophysics to life sciences [1]. The radioactive nuclides are produced via spallation, fission, and fragmentation reactions originating from a 1.4 GeV proton beam that impinges on specifically designed thick targets. For more than 50 years, the ISOLDE facility at CERN [2] has been delivering radioactive ion beams to users from all around the world, extending the knowledge of radionuclides for a large panel of applications. To extend the benefits of this knowledge to societal applications,

representative picture of the ISOLDE targets. More details on the target design and operation can be found in [6][7][8]. Different materials can be used for the target core (as UC2-Cx, ThC2-Cx, Pb, Ta, etc.) depending on the experimental needs. As of January 2021, the most used target material (70-80 % of all targets) is the uranium carbide (UC2-Cx) simply referred to as UC2C. A target can be expected to decommissioned after having received about 5.0×10^{18} protons over an irradiation period spanning from a few days to a couple of weeks depending on the beam availability and the operation schedule.

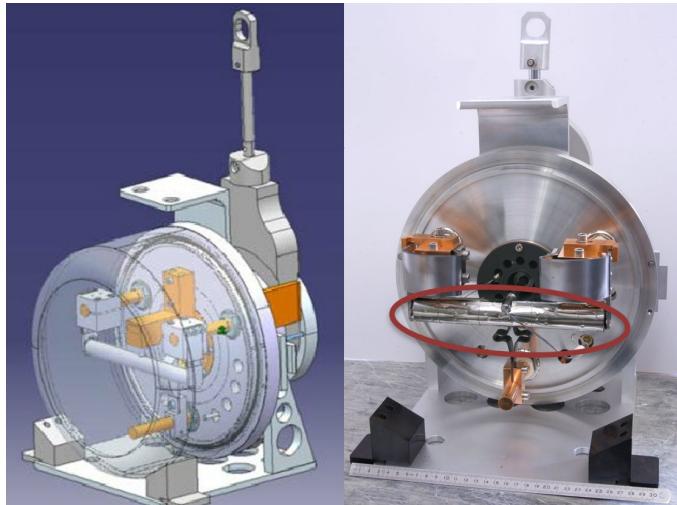


Fig. 1. 3D model rendering and picture of an ISOLDE target assembly. The so-called casserole, partially transparent in the drawing, was removed in the picture on the right to show the target core (circled in red) and other auxiliary components.

The MEDICIS experimental facility (MEDical Isotopes Collected from ISOLDE) is designed to produce radionuclides for medical applications. The MEDICIS operation is driven by the ISOLDE facility and, as for the latter, it receives protons from the Proton Synchrotron Booster with the same energy and intensity. As of February 2021, the MEDICIS irradiation station is located downstream, with respect to the beam traveling direction, of the ISOLDE High-Resolution Separator station (HRS). Thus, a MEDICIS target being placed downstream of an ISOLDE target, receives the fraction of primary protons which have not interacted with the ISOLDE target plus the secondary particles produced by the beam interaction with the latter [9].

For this paper, only Tantalum targets are considered even though the nuclide inventory calculations are performed for a wide variety of target core materials. The latter will be the subject of future publications.

B. Radionuclide Inventory and Scaling Factor Estimation Method

1) FLUKA modeling and Particle Fluence Spectra

To estimate the nuclide inventory for the multiple irradiation configurations and conditions, to which the ISOLDE and MEDICIS targets can be exposed, a multiple-step scheme is used. First, with the Monte Carlo particle transport code FLUKA [10] [11] [12], different irradiation configurations are

modeled and simulated. An irradiation configuration is defined by combinations of the following parameters:

- The ISOLDE and MEDICIS target core materials,
- The type of target placed upstream of the MEDICIS target (for MEDICIS targets only),
- The use of the so-called neutron converter [8].

The aim is to calculate, using FLUKA, the proton, neutron, positive and negative pion fluence spectra produced by the interaction of the primary proton beam with the target material Tantalum (Ta) given the target area geometry for each of the possible irradiation configurations. For the characterization of the core materials, it is also necessary to score the photon fluence spectra inside the core material. These irradiation configurations are used to obtain two sets of particle fluence spectra:

- A first set representative for all the structural materials (e.g. aluminum, copper, stainless steel, etc.) calculated in a region outside the target core,
- A second set, including the photon fluence, for the Tantalum core material calculated inside the target core.

Once the particle fluence spectra are calculated for all the relevant irradiation configurations, the material activation calculations are performed with ActiWiz Creator [13]. ActiWiz Creator, hereinafter simply ActiWiz, is an analytical software whose core functionality allows calculating the nuclide inventory of a user-defined material given the following parameters:

- Neutron, proton, photon, positive and negative pion fluence spectra,
- Material elemental composition and density,
- Irradiation and cooling times,
- Beam intensity expressed in protons per second.

The ActiWiz analytical calculations are less computationally demanding compared with the Monte Carlo FLUKA simulations. It is, therefore, possible to perform a large set of calculations, covering all possible irradiation scenarios. In particular, for every irradiation configuration simulated in FLUKA, we calculate the residual radionuclide inventories for all the relevant cooling times and for any given activated material. Finally, the material-specific nuclide inventories obtained for all the core and structural materials, given all the combinations of irradiation configurations and scenarios are statistically analyzed to extract relevant information such as specific activities.

A simplified FLUKA model of the ISOLDE-MEDICIS target unit and its surroundings was produced. As can be seen from Figure 2, the model consists of an inner cylinder containing the target core material (Ta). The target core material is pure tantalum, with a density of 5.0 g/cm^3 . The latter is surrounded by a second cylinder which represents the target unit structure (i.e. casserole, target flange, etc.). The distance between the inner and the outer cylinder is representative of the average distance between the target core and the other structures of the target unit. The geometry dimensions are presented in Table I which includes the distance from the

ISOLDE and MEDICIS target that is placed upstream (see Figure 3). The target unit thus modeled is placed inside a concrete box, whose walls are 200 cm thick, which models the walls, floor, and ceiling of the irradiation station (see Figure 3). The position of the target model inside the concrete box is representative of the actual position of the target unit in the irradiation station. The concrete walls and the beam dump are expected to affect the particle fluence spectra inside the target unit by means of back-scattered radiation (e.g. thermal neutrons back-scattered into the target from the concrete walls). Therefore, it was decided to include them in the model.

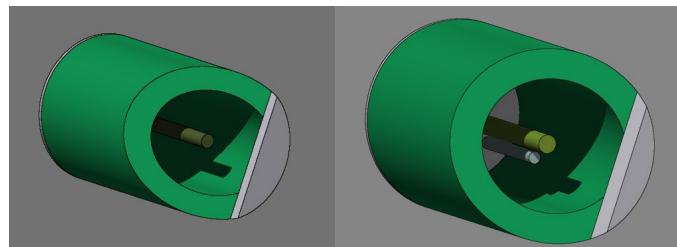


Fig. 2. (Left) 3D plot of the FLUKA model for an ISOLDE-MEDICIS target unit showing the target container, the outer cylinder, and its base plates. (Right) 3D plot of the FLUKA model for an ISOLDE target featuring a neutron converter placed below the target container.

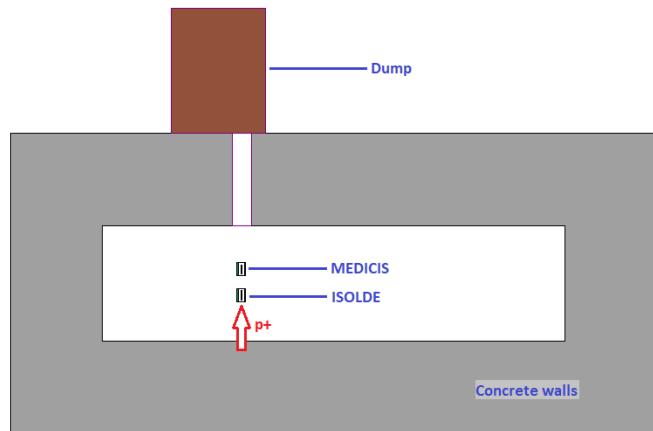


Fig. 3. Top view of the FLUKA model showing the ISOLDE and MEDICIS targets surrounded by the concrete walls with the opening to the beam dump.

TABLE I
MAIN DIMENSIONS OF THE FLUKA MODELS USED FOR THE FLUENCE SPECTRA CALCULATIONS. THE “DIST FROM TARGET CONTAINER” IS THE DISTANCE BETWEEN THE AXIS OF THE TARGET CONTAINER CYLINDER AND THE AXIS OF THE NEUTRON CONVERTER CYLINDER. THE “DIST. FROM ISOLDE TARGET” IS THE DISTANCE BETWEEN THE CENTRE OF THE ISOLDE TARGET CONTAINER AND THE MEDICIS TARGET CONTAINER.

| Element | Dimension | Value [cm] |
|----------------------|-------------|------------|
| Target container | Radius | 0.9 |
| | Height | 20 |
| Outer cylinder | Inn. radius | 6.45 |
| | Out. radius | 9.32 |
| Out. cylinder plates | Height | 29 |
| | Radius | 9.32 |
| | Height | 0.5 |

| | | |
|-------------------|---------------------------------|------|
| Neutron converter | Radius | 0.6 |
| | Height | 12.5 |
| | Dist. from the target container | 1.75 |
| MEDICIS target | Dist. From ISOLDE target | 55.5 |

The base plates of the outer cylinder representing the target unit are made of aluminum as the actual target casserole. Whereas the body of the outer cylinder is made of a dummy material obtained by combining the four structural materials which dominate the total mass of the target unit. These are aluminum alloys (AW 6082 and AW 5083), stainless steel alloys (304 and 316), copper (CU ETP C11000) and pure tantalum. Hence, for the elemental compositions of the structural material, a representative composition was obtained by estimating the Main structural materials found in an ISOLDE-MEDICIS target (see Table II).

In one configuration, the proton beam energy is set to 1.4 GeV. The beam impinges on the target core after having passed through the aluminum base plate of the outer cylinder. Otherwise, the beam impacts on the neutron converter, after having passed through the aluminum base plate, for the corresponding irradiation configurations.

As stated previously, the fluence spectra for protons, neutrons, positive and negative pions are scored in FLUKA simulations. The scoring regions are the target container (i.e. inner cylinder in Figure 2 Left) and the outer cylinder representative of the unit structural materials (i.e. outer cylinder in Figure 2).

TABLE II
ELEMENTAL COMPOSITION OF THE DUMMY MATERIAL USED FOR THE FLUENCE SPECTRA CALCULATION AND REPRESENTATIVE OF THE MAIN STRUCTURAL MATERIALS FOUND IN THE ISOLDE-MEDICIS TARGET

| Element | Mass fraction | Element | Mass fraction |
|-----------|---------------|-------------|---------------|
| Aluminum | 5.95E-01 | Magnesium | 5.57E-03 |
| Copper | 1.93E-01 | Zinc | 1.24E-03 |
| Iron | 8.84E-02 | Titanium | 6.19E-04 |
| Tantalum | 6.39E-02 | Nitrogen | 1.25E-04 |
| Chromium | 2.54E-02 | Phosphorous | 5.64E-05 |
| Nickel | 1.25E-02 | Oxygen | 4.33E-05 |
| Silicon | 7.13E-03 | Carbon | 3.76E-05 |
| Manganese | 6.84E-03 | Sulphur | 3.76E-05 |

Structural Material Irradiation Configurations

For the radiological characterization of the ISOLDE structural materials the following irradiation configurations are considered:

- Direct irradiation of a tantalum target core, and
- Indirect irradiation of a tantalum target core through the neutron converter.

The irradiation station for MEDICIS targets is located downstream of the ISOLDE irradiation station with respect to

the beam travelling direction. Therefore, the MEDICIS targets can either receive an unperturbed proton beam (no ISOLDE target located upstream) or a proton beam which has interacted with an ISOLDE target located upstream plus the produced secondary radiation (i.e. neutron, secondary protons, and pions). In the former case, the MEDICIS target irradiation configuration is equivalent to an ISOLDE target, whereas, in the latter, the MEDICIS target irradiation depends upon the type of ISOLDE target located upstream. Therefore, for the radiological characterization of MEDICIS structural materials the following irradiation configurations are considered:

- Indirect irradiation of a tantalum target core through a UC2C ISOLDE core, and
- Indirect irradiation of a tantalum target core through the ISOLDE neutron converter.

Target containers, made of tantalum, are used to accommodate the target material that interacts directly with the proton beam. The target container is located only a few millimeters away from the beam interaction point (i.e. from the target core). Dedicated FLUKA simulations and ActiWiz Creator calculations showed that the particle fluence spectra scored inside the tantalum core can be used to produce conservative and representative estimates of the container activation. Therefore, the activity of the target container is calculated with the particle fluence spectra scored inside a tantalum core for the irradiation configurations described above.

Core Material Irradiation Configurations

The activation of the ISOLDE and MEDICIS core materials is largely dominated by the nuclear reactions induced by the primary proton beam. The only exception is when the proton beam impinges on the neutron converter leading to the production of a higher neutron fluence per primary proton. Therefore, for the radiological characterization of the core materials, the following two irradiation configurations are used for both the ISOLDE and MEDICIS targets:

- Direct irradiation of the core material: this configuration is representative of the direct irradiation of any tantalum core material in the ISOLDE and MEDICIS targets,
- Indirect irradiation of the core material through the neutron converter: this irradiation is representative of the indirect irradiation of the tantalum core material in the ISOLDE and MEDICIS targets.

2) Activation Calculations With ActiWiz Creator

A necessary set of input parameters is required by the tool ActiWiz Creator to calculate the nuclide inventories for the considered core and structural materials given the particle fluence spectra obtained with FLUKA. The material compositions and densities of the structural and core materials used to perform the ActiWiz Creator calculations are provided hereafter. The ISOLDE and MEDICIS targets are irradiated for several days depending on the proton beam availability. As the cooling times are typically in the order of years, the irradiation time can be considered as instantaneous (i.e. 1 second) for

activation studies. The total number of protons on target (POT) generally does not exceed 5.0E+18. The cooling times range from 3 to 30 years in one-year steps.

3) Scaling Factor Calculation

A scaling factor (SF) is the ratio of the activity of a nuclide that cannot be measured via gamma spectroscopy (e.g. pure beta emitter) and one that, if present in the waste, is systematically and readily measured (e.g. Co-60, Na-22). The former are categorized as difficult-to-measure (i.e. DTM) depending on their radiological characteristics, whereas the latter is the so-called key nuclides (i.e. KN) belonging to the list of easy-to-measure isotopes (i.e. ETM).

Therefore, by measuring the activity of the ETMs via gamma spectrometry, one can estimate the activity of the DTMs by multiplying the activity of the selected key nuclide by the corresponding scaling factor.

Scaling factors are calculated based on the expected activities in a material or waste package (i.e. mixture of waste materials) upon verification of the existence of a mathematical correlation between the activity levels of the DTMs and the chosen key nuclide

4) Key Nuclide Selection

Given the radionuclide inventories and activity levels discussed previously, the first step towards the calculation of the scaling factors consists of selecting a key nuclide for each material. Generally, a key nuclide shall satisfy the following criteria [14] :

- It can be detected by gamma spectrometry,
- It is expected to have an activity higher than its detection limit (also known as Minimum Detectable Activity, MDA),
- Its activity value is correlated with the activity of the DTM of interest,
- It has a relatively long half-life (e.g. years rather than days),
- It is exclusively produced by the activation of the considered material, if possible.

C. Hot Cell dismantling and Drum packaging

The targets are dismantled in a dedicated hot cell (HC) (see Figure 4 Left). It enables the disassembly without cutting or grinding. The choice to perform such a process in a hot cell is due to the targets high dose rate (up to 2 mSv/h at contact) and the associated contamination risk. A hot cell offers a two-way protection by:

- Shielding from radiation with a 150 mm lead shielding all around and 150 mm thick glass, and
- Confining contamination with a leak-tight envelope and a lower atmospheric pressure compared surrounding volumes.

The mechanical telemanipulators are the main instruments to action anything inside the HC.

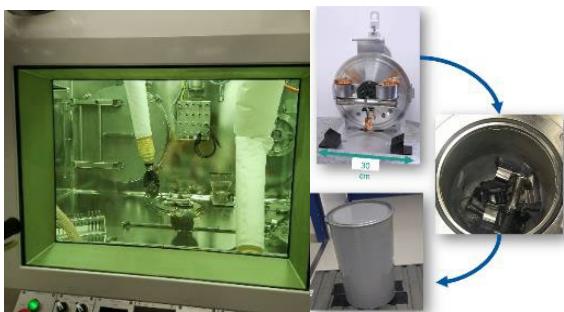


Fig. 4. (Left) view of the hot cell at the ISOLDE facility used to dismantle and package the targets into drums. (right) a process showing the packaging of the target aluminum containers into a 107 L drum.

The dismantled targets are then packaged into three waste drums belonging to the following categories as shown in the right of Figure 4:

- Structural material in 107 L drums: Aluminum,
- Target material in 21 L drums: Tantalum,
- Other structural materials in 21 L drums.

D. Gamma Spectrometry (GS) Setup

GS is a widely deployed non-destructive assay technique at CERN to quantify the residual activity of gamma emitters or ETM nuclides in various items, ranging from small-volume samples in a laboratory to large objects such as unitary radioactive waste packages. For this study, we use high energy resolution germanium detectors Falcon 5000 from Mirion technologies (Canberra) as shown in Figure 5. To generate the efficiency calibration curves, one needs to know several geometry parameters including the dimensions of the material, the distance between the detector and its composition (material information). ISOCS (In Situ Counting Object System) and LabSOCS (Laboratory Sourceless Calibration Software) from Mirion Technologies (Canberra) are applied in the laboratory for creating efficiency calibrations of good quality without using radioactive standards at the laboratory [16] [17] [18] [19].



Fig. 5. View of the gamma spectrometry setup.

III. RESULTS AND DISCUSSION

A. Radionuclide Inventory and Scaling Factor

For the purpose of this study, it was decided to choose the third statistical quartile of the activity ratio distribution as the retained scaling factor value. This choice is considered robust as it does not require any specific distribution assumption of the SF (e.g. log-normal) and is less sensitive to data outliers. Figure 6 shows a third quartile example obtained for the SF distribution for the ratio H-3/Na-22. In this case, H-3 is the DTM while Na-22 nuclide is considered as the KN.

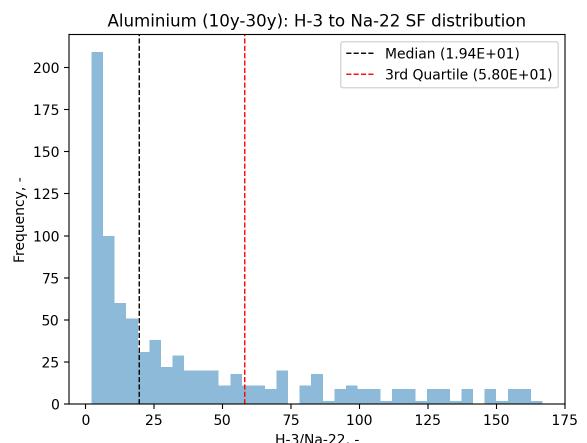


Fig. 6. Histogram of the H-3/Na-22 activity ratios for the ISOLDE aluminum alloys for the cooling time period 10y-30y.

The selected key nuclides for the materials considered in this study are listed in Table III. The materials EN AW 6082 and EN AW 5083 are grouped in “aluminum alloys”, and AISI 304, AISI 304L, AISI 316, S235JR in “steel alloys”.

TABLE III
LIST OF SELECTED KEY NUCLIDES.

| Material | Key nuclide |
|-----------------|-------------|
| Al2O3 | Na-22 |
| Aluminum alloys | Na-22 |
| Brass | Co-60 |
| CU-ETP-C11000 | Co-60 |
| Molybdenum | Nb-94 |
| NiFe42 | Co-60 |
| Steel alloys | Co-60 |
| Tantalum | Hf-178n |
| Tungsten | Co-60 |

The scaling factors for the tantalum are defined as the mass-weighted mean of the third quartile of the distributions obtained for the structural materials, and the third quartile of the distributions obtained for the core material. Given the material content allowed in each waste category, it is possible to produce three sets of combined scaling factors for each waste category. That is, one set per cooling time period (i.e. 3y-10y, 10y-30y,

and 3y-30y) per waste category. The combined scaling factors are obtained by taking the highest scaling factor for each DTM expected to be found in each waste category for a given key nuclide. For instance, the waste packages belonging to the category “Structural material: aluminum” are expected to contain the key nuclide Na-22. This key nuclide is related to the presence of both Al_2O_3 and aluminum alloys. Since the DTM nuclide H-3 is produced by the activation of both materials, two values of the H-3/Na-22 scaling factor are available – one for Al_2O_3 and one for the aluminum alloys. A conservative estimate of the H-3 activity in the waste is obtained by selecting the highest scaling factor between the aforementioned values. Similarly, if a DTM is related to two or more key nuclides found in the waste category, the DTM total activity shall be estimated by summing the values derived from each scaling factor. For example, the waste packages belonging to the category “Structural material: aluminum” are expected to contain the key nuclides Na-22 and Co-60. The former is due to the presence of Al_2O_3 and aluminum alloys, whereas the latter is due to steel alloys. For instance, the DTM nuclide Cl-36 is produced by the activation of both the aluminum and steel alloys. Hence, the Cl-36 activity contained in a waste package shall be calculated by summing the activity estimate based on the Cl-36/Na-22 scaling factor (aluminum alloys), and the one estimated via the Cl-36/Co-60 scaling factor (steel alloys). The scaling Factor values for 10 to 30 years decay time that are above 2% are shown in Figure 7.

| Nuclides | | Na-22 | ↓ | Nuclide | Co-60 | ↓ |
|----------|----------|-------|---|---------|----------|---|
| H-3 | 1.44E+02 | | | H-3 | 1.50E+04 | |
| C-14 | 4.26E-01 | | | Ta-179 | 6.72E+02 | |
| Ti-44 | 2.32E-02 | | | Hf-172 | 2.05E+02 | |
| Al-26 | 1.05E-02 | | | Pm-145 | 1.51E+02 | |
| | | | | Lu-173 | 4.00E+01 | |
| | | | | Lu-174 | 3.46E+01 | |
| | | | | Ba-133 | 1.65E+01 | |
| | | | | Ho-163 | 1.24E+01 | |
| | | | | Fe-55 | 1.08E+01 | |
| | | | | Ni-63 | 7.05E+00 | |
| | | | | Kr-85 | 2.13E+00 | |
| | | | | Eu-150 | 7.11E-01 | |
| | | | | Ar-42 | 4.96E-01 | |
| | | | | Cd-113m | 3.91E-01 | |
| | | | | Eu-152 | 2.56E-01 | |
| | | | | C-14 | 2.44E-01 | |
| | | | | Ti-44 | 2.26E-01 | |
| | | | | Rh-101 | 2.08E-01 | |
| | | | | Ar-39 | 1.83E-01 | |
| | | | | Rh-102 | 1.62E-01 | |
| | | | | Nb-93m | 1.12E-01 | |
| | | | | Pm-146 | 1.07E-01 | |
| | | | | Ag-108m | 8.19E-02 | |
| | | | | Sm-145 | 2.79E-02 | |
| | | | | Pt-193 | 2.04E-02 | |
| | | | | Tl-204 | 1.01E-02 | |
| Nuclide | | Nb-94 | ↓ | Nuclide | Hf-178m | ↓ |
| H-3 | 7.34E+02 | | | H-3 | 1.11E+02 | |
| Nb-93m | 3.88E+02 | | | Pm-145 | 5.92E+00 | |
| Nb-91 | 1.60E+02 | | | Gd-148 | 2.75E+00 | |
| Mo-93 | 2.91E+01 | | | Ba-133 | 2.49E+00 | |
| Tc-99 | 7.78E-01 | | | Tb-157 | 1.99E+00 | |
| Sr-90 | 6.63E-01 | | | Ta-179 | 5.96E-01 | |
| Kr-85 | 1.39E-01 | | | Hf-172 | 5.78E-01 | |
| Kr-81 | 1.93E-02 | | | Lu-174 | 5.17E-01 | |
| | | | | Lu-173 | 1.10E-01 | |
| | | | | Ho-163 | 6.23E-02 | |
| | | | | Ni-63 | 4.22E-02 | |
| | | | | Nb-93m | 3.13E-02 | |
| | | | | Ar-42 | 1.66E-02 | |
| | | | | Rh-101 | 1.56E-02 | |
| | | | | Eu-150 | 1.31E-02 | |
| | | | | Ar-39 | 1.13E-02 | |
| | | | | Fe-55 | 1.09E-02 | |
| | | | | Kr-85 | 1.08E-02 | |

Fig. 7. Scaling Factor values for 10 to 30 years decay time. Due to the large size of the tables, only SF values above 2% are included.

B. ETM nuclides activity Estimation

The GS technique presents several challenges during both the acquisition and the analysis steps. The challenges are summarized when establishing the following:

- The nuclide library,
- The γ -spectrometry counting time and the allowed data acquisition dead time limit, and
- The best geometry model for efficiency calibration needed for activity calculations.

Regarding the dead time, the performed studies show that there

is no systematic activity bias beyond 10% if the GS acquisition dead time was maintained below a value of approximately 10%. A typical GS spectrum is shown in Figure 8 and a typical GS geometry model, used for efficiency calibrations, is shown in Figure 9.

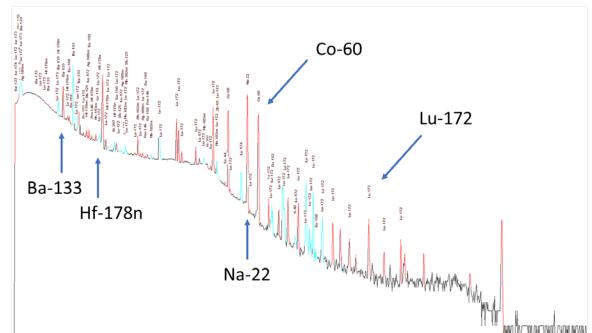


Fig. 8. A typical GS spectrum for the Ta 21 L drum.

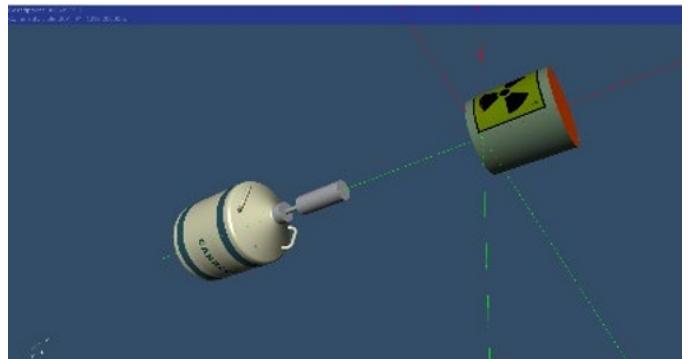


Fig. 9. A typical GS ISOCS geometry model for the Ta 21 L drum.

To validate the ETM estimation using GS, two targets ID#239 and #308 are dismantled in the HC. During this process, targets have been separated into three drum categories for disposal (Structural material: aluminum 107 L drum, target material: tantalum 21 L drum, and other structural materials 21 L drum). The sum of activities obtained for the target ID#239 and #308 (target unit modeling) and for the three drums (Al-Drum, Other-Drum and Ta-Drum modeling) are compared. The drums and targets activity ratio illustrated in Figure 10 is comprised between 0.69 and 1.48. Therefore, for targets ID#239 and #308, the measured activity ratio is within $\pm 50\%$, which is consistent with the estimated activity ratio uncertainty at $k=2$ (see Figure 9).

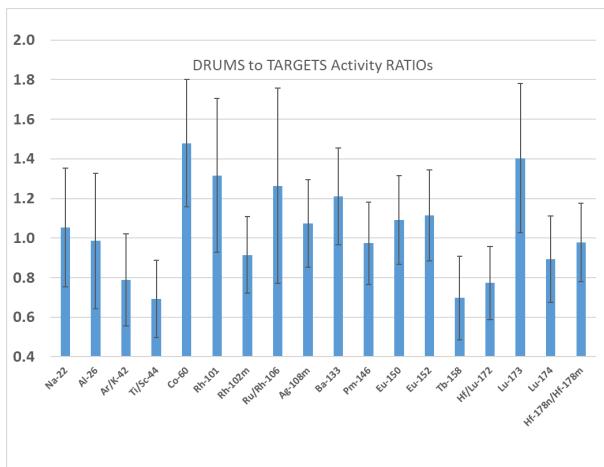


Fig. 10. Drums to targets activity ratio and error bars (the uncertainties are reported at 1σ).

IV. CONCLUSIONS

ISOLDE and MEDICIS are two experimental facilities located at CERN where fixed targets are irradiated by protons beam to produce radioactive ion beams. Currently, over 400 targets are stored at CERN (as of 15/08/2023) and, on average, 30 targets are irradiated every year. All these targets will be dismantled and disposed of as radioactive waste in a dedicated waste repository in Switzerland.

In this study, a three-step scheme was developed to estimate the nuclide inventories resulting from the activation of the core and structural materials of which ISOLDE and MEDICIS targets are made. The steps can be summarized by:

- Predicting the radionuclide inventory and associated DTM scaling factors taking into account the targets irradiation conditions and material elemental compositions,
- Dismantling and conditioning the waste into the various drum types, and
- Performing the Gamma Spectrometry analysis to estimate the ETM radionuclides and deduce the activity values of the other DTM nuclides.

This study describes the methodology used to estimate the complete radionuclide inventory and activity levels of present and future targets based on extensive analytical calculations and the associated non-destructive assay (NDA) technique by evaluating the counting conditions and establishing the efficiency calibration curves using ISOCS (In Situ Counting Object Software) from Mirion Technologies (Canberra). The study also compares the ETM activities obtained by the NDA technique before and after packaging the targets following the segregation step into the three material types. This paper addresses each of these challenges and offers technical solutions based on computational codes, statistical techniques and state of the NDA technique using high energy resolution gamma spectrometry. We believe that the methodology here presented can be applied to activation and characterization studies that are characterized by a relatively large number of

possible activation scenarios and input parameters.

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REFERENCES

- [1] Catherall, R. et al. The ISOLDE facility. *Journal of Physics G: Nuclear and Particle Physics* **44**, 094002 (2017).
- [2] Borge, M. J. & Jonson, B. ISOLDE past, present and future. *Journal of Physics G: Nuclear and Particle Physics* **44**, 044011 (2017).
- [3] Augusto, R. M. d. S. et al. CERN-MEDICIS (medical isotopes collected from ISOLDE): a new facility. *Applied Sciences* **4**, 265–281(2014).
- [4] Duchemin, C. et al. CERN-MEDICIS: A review since commissioning in 2017. *Frontiers in Medicine* **8** (2021).
- [5] Accord entre le Conseil Fédéral Suisse, le Gouvernement de la République Française, et l'Organisation Européenne pour la Recherche Nucléaire relatif à la Protection contre les rayonnements ionisants et à la Sécurité des Installations de l'Organisation Européenne pour la Recherche Nucléaire, conclu le 15 novembre 2010, entré en vigueur par échange de notes le 16 septembre 2011. Number RS 0.814.592.2, Recueil O-ciel n. 34 du 23 août 2011, page 3825. Décret n. 2011-1024 du 24 août 2011, JORF n. 0199 du 28 août 2011 page 14594. 2011.
- [6] M. Brice, An ISOLDE target unit. Cible ISOLDE, CERN-EX-0204023.
- [7] T. Stora, Radioactive Ion Sources. CAS-CERN Accelerator School: Ion Sources - Proceedings. 10.5170/CERN-2013-007.331 (2014).
- [8] A. Gottberg et. al., Experimental tests of an advanced proton-to-neutron converter at ISOLDE-CERN, *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms*, Volume 336, 2014, Pages 143-148, ISSN 0168-583X, <https://doi.org/10.1016/j.nimb.2014.04.026>.
- [9] C. Duchemin et al., CERN-MEDICIS: A Unique Facility for the Production of Nonconventional Radionuclides for the Medical Research, 11th International Particle Accelerator Conference (2020).
- [10] CERN,online:<https://fluka.cern/>, (Last accessed 15 August 2023).
- [11] Ahidida, C. et al. New capabilities of the FLUKA multi-purpose code. *Front.Phys.9,14* (2022).
- [12] G. Battistoni et al., Overview of the FLUKA code, *Annals of Nuclear Energy* **82**, 10-18 (2015).
- [13] H. Vincke and C. Theis, Actiwiz 3 - an overview of the latest developments and their application, *Journal of Physics Conference Series*, no. 1046(1):012007 (2018).
- [14] IAEA, Determination and use of scaling factors for waste characterisation in nuclear power plants, IAEA Nuclear Energy Series No. NW-T-1.18 (2009).
- [15] B. Zaffora et al., Radionuclide inventory and scaling factors of metallic, TFA radioactive waste, EDMS 1868767 v.1.2 (2019).
- [16] Patrycja Dyrz, Thomas Frosio, Nabil Menaa, Matteo Magistris, and Chris Theis. Qualification of the activities measured by gamma spectrometry on unitary items of intermediate-level radioactive waste from particle accelerators. *Appl. Radiat. Isot.*, 167:109431, 2021. doi:10.1016/j.apradiso.2020.109431
- [17] F. Bronson. ISOCS, a Laboratory Quality Ge Gamma Spectroscopy system you can take to the Source for Immediate High Quality Results. *Proceedings of the Rapid Radioactivity Measurements in Emergency and Routine Situations Conference*, UK, October 1997.
- [18] N. Menaa, A. Bosko, F. Bronson, R. Venkataraman, W. R. Russ, W. Mueller, V. Nizhnik, and L. Mirolo. Mathematical efficiency calibration with uncertain source geometries using smart optimization. In 2011 2nd International Conference on Advancements in Nuclear Instrumentation, Measurement Methods and their Applications, pages 17, 2011. doi: 10.1109/ANIMMA.2011.6172913.
- [19] Thomas Frosio, Nabil Menaa, Philippe Bertreix, Maeva Rimlinger, and Chris Theis. A novel technique for the optimization and reduction of gamma spectroscopy geometry uncertainties. *Applied Radiation and Isotopes*, 156:108953, 2020. ISSN 0969-8043. doi: <https://doi.org/10.1016/j.apradiso.2019.108953>. URL <https://www.sciencedirect.com/science/article/pii/S0969804319306852>.