

PRELIMINARY ASSESSMENT OF THE RADIONUCLIDES INVENTORY IN IRRADIATED AlMg₃ ALLOY

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The irradiated aluminium that will result from TRIGA decommissioning (foreseen to be SHUT-DOWN in 2035) is considered a problematic radioactive waste category for which it will be necessary to establish an appropriate management strategy. The paper presents the results obtained by MCNP and ORIGEN computer code calculations performed to estimate the radionuclides inventory in AlMg₃ alloy samples irradiated in controlled conditions in the TRIGA reactor. The chemical composition of the sample was determined by SEM-EDS, EDXRF, and ICP-OS measurements as it is a necessary parameter in the accuracy of computer code modeling. The results obtained highlighted the presence of ⁶⁰Co, ⁶⁵Zn, ³H, ⁶³Ni, ⁵⁵Fe, ⁵⁹Fe, ⁵⁴Mn radionuclides in the AlMg₃ alloy irradiated samples. This preliminary inventory data will be used to develop the characterization strategy of the aluminium waste that will be generated by TRIGA decommissioning.

Keywords: MCNP6, ORIGEN-S, decommissioning, radionuclides inventory.

1. Introduction

At the end of their life, all nuclear facilities have to be decommissioned and some of them will be released from the regulatory control. The decommissioning strategy involves preliminary and detailed planning activities, cost estimation, decontamination, and dismantling techniques, and also, plans for the safe management of the decommissioning waste [1].

Establishing all the technical and organizational activities requires an estimation of the amount and the type of radioactivity contained in the materials surrounding the reactor core. The construction materials such as aluminium, stainless steel, concrete, lead, carbon steel, iron, boron carbide, etc. are some of those radioactive wastes generated by the decommissioning activities containing

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small quantities of radionuclides generated mainly by impurities activation and for which the inventory must be determined.

TRIGA reactor in operation since 1980 at the Institute for Nuclear Research Pitesti site consists of two active cores and it is mainly used for research, radioisotope production, and irradiation testing of nuclear fuel and structural materials.

TRIGA reactor is foreseen to be shut down in 2035. For this reason, preliminary studies to estimate the amount of decommissioning waste and the radionuclides inventory are required for the preparation and planning of all the decommissioning activities and, for the safe management of the radioactive waste generated. Currently, in Romania the institutional low and intermediate, short-lived waste are disposed of in Băița Bihor National Repository (DNDR). Except for the waste that contains long-lived radionuclides in amount exceeding the waste acceptance criteria (WAC) for DNDR, the short-lived radioactive waste resulted from TRIGA reactor decommissioning are planned to be treated and conditioned accordingly with the WAC for DNDR.

The radionuclides inventory can be estimated based on theoretical calculations taking into account the sample geometry, the material composition, and the irradiated history, and by experimental measurements performed on different samples from the interest regions [2].

Because the TRIGA reactor is still in operation, the inventory estimation can be performed only by theoretical calculations using modeling codes correlated with the experimental tests carried out on irradiated materials similar to those that will result from the decommissioning activities.

The irradiated aluminium that will result from TRIGA decommissioning is considered a problematic radioactive waste category due to its reactivity with the currently used conditioning matrix based on Portland cement.

In the TRIGA reactor, the aluminium alloy is used as construction material for the reactor tank, beam tubes, grids support and shroud of fuel clusters, parts of the transfer channel from the reactor pool to the hot cells facility, shroud assembly of the control rods and the racks used for irradiated fuel storage. It is assumed that around 6 tons of irradiated aluminium will result from TRIGA decommissioning activities [3].

Many authors in their research studies related to nuclear facilities decommissioning applied MCNP and ORIGEN codes to estimate the activity inventory in the structural components of a research reactor [4-10] as biological shield concrete [5], steel and aluminium [6], graphite reflectors, flue gas [7], spent nuclear fuel [8]. Also, for the initial estimation of the radionuclide inventory in the VVR-S reactor block a combination of DORT and ORIGEN codes were used [9, 10].

One of the most important parameters in the computer codes calculation is the accuracy of chemical composition data of the material of interest because almost all the radioactivity is due to the neutron activation of impurities in the material.

Techniques, such as Induced Coupled Plasma Mass Spectrometry (ICP-MS), inductively coupled plasma emission spectroscopy (ICP-OES) and Flame Atomic Absorption Spectroscopy (FAAS) are used to determine the metallic species concentration (such as Zn, Fe, Ni, Cu, Pb and Cr) in solutions obtained by material dissolution using specific protocols [11÷13]. Techniques such as Energy Dispersive X-Ray Fluorescence Spectrometry (EDXRF), Scanning Electron Microscopy and Energy-Dispersive X-ray Spectroscopy (SEM-EDS) analysis have been used to quantify the major compounds by direct analyses of the solid samples [14÷17].

The purpose of this study was to estimate the radionuclides inventory in the activated aluminium alloy (AlMg₃) irradiated in controlled conditions in the TRIGA reactor to obtain preliminary information on the radionuclides inventory in the aluminium waste generated by TRIGA decommissioning. To accomplish the goal of this study an AlMg₃ alloy sample was irradiated in a TRIGA reactor for 691.148 h at 12.117 MW and to evaluate the radionuclides inventory the MCNP6 and ORIGEN-S codes were used. The chemical composition of AlMg₃ alloy was determined by SEM-EDS, EDXRF, and by ICP-OES. The aluminium composition is an important parameter in the modeling code calculation accuracy. The results obtained will be used as preliminary information for developing a characterization plan for the aluminium waste that will be generated by TRIGA decommissioning.

Future studies will focus on the experimental tests carried out on the same irradiated aluminium sample to confirm the modeling results. As impurities are not uniformly distributed in the aluminium alloy, uncertainty tests using the minimum and maximum values of the chemical composition measured by ICP-OES will be performed using MCNP6 and ORIGEN-S codes.

2. Methods

2.1. Materials

Aluminium sampling from the reactor active zone is impossible as the TRIGA reactor is still in operation and, to estimate the radionuclides inventory for this type of material, a cylindrical AlMg₃ bar of 250x10x6 mm and density of 2.7 g/cm³ was cut from a larger AlMg₃ bar and irradiated in the reactor.

The MCNP6 input was created taking into account the physical and chemical characteristics of the irradiated aluminium bar. Small representative samples were cut from the same inactive AlMg₃ alloy to perform its characterization. The inactive AlMg₃ alloy used has similar physical and chemical characteristics as the aluminium alloy used as a structural material in the TRIGA reactor. Considering that this type of aluminium alloy contains more than 95% Al

[18], the main challenge in establishing with adequate accuracy its chemical composition is to measure the impurities content. Four small aluminium samples were dissolved and the resulted solutions were prepared according to the ICP OES measurements protocol to determine the impurities in the AlMg₃ alloy.

2.2. Characterization methods

In the first step of our study, the SEM-EDS and EDXRF analyses were performed to obtain preliminary data regarding the chemical composition, homogeneity and distribution of the impurities in the AlMg₃ alloy sample.

SEM-EDS and EDXRF are nondestructive characterization techniques that provide chemical elemental analysis of the material surface. By SEM technique a specific region of the sample is scanned using an electron beam and the EDS module can perform point scans or mapping analyzes on the evaluated region [12].

EDXRF technique is based on the interaction of X-rays with the material to determine the chemical composition. SEM-EDS measurements were carried out using a HITACHI SU5000 scanning electron microscope combined with an energy dispersive detector (EDS). EDXRF analyses were performed using a SPECTRO MIDEX M spectrometer enabling rapid spot analysis of unidentified samples, which allows the determination of any elements from sodium to uranium. The impurities contained in the AlMg₃ alloy sample were measured by the ICP-OES technique that is considered with less interferences, detection limit in the ppb region for some elements in the sample matrix, and optical spectrum in the range of 165 - 800 nm [17]. The ICP-OES measurements were performed using an iCAP 6500 spectrometer produced by ThermoScientific, with dual plasma viewing (axial and radial), and the measurement was based on the spontaneous emission of photons from atoms and ions excited by absorbing energy from plasma. The concentration of the element in the analyzed sample is directly proportional to the number of photons of a certain wavelength [15].

The calibration standards of the emission spectrometer were prepared in 2% HNO₃ in different concentration ranges: between 3 and 15 mg/L for Al, Fe, Zn, Si, Mg, Mn, Cr, and Cu, between 0.1 and 1 mg/L for Co, Ni and Sc, and 0.1 - 0.3 mg/L for Li. The aluminium samples analyzed by ICP-OES were dissolved in a mixture of concentrate acids (HNO₃ and HCl) at room temperature. The solutions were evaporated to wet salts and dissolved in 50 ml ultrapure 2% HNO₃. To minimize the matrix effect due to the presence of Al in high concentrations in the AlMg₃ matrix, an aspect that can influence the accuracy of ICP-OES measurements, in the calibration standards 300 mg/L of Al mono-element ICP standard solution was added. Also, 5 mg/L of Y mono-element ICP standard solution was added both in the calibration standards and in the samples to be analyzed [15]. As an internal standard, yttrium is used to adjust for chemical and physical interference that may have an impact on the accuracy of the results.

2.3. Calculation Methods

To evaluate the amount and the types of radionuclides accumulated in the irradiated aluminium bar, a combination of computer codes such as MCNP6, CSAS1X, COUPLE modules, and ORIGEN-S were used.

The MCNP6 is a general-purpose, constant-energy, general-geometry, time-dependent Monte Carlo radiation-transport code, designed to track different types of particles such as neutrons, photons, and electrons, in a wide energy range. The neutron energies range from 10^{-11} MeV to 20 MeV for all isotopes and up to 150 MeV for some isotopes, the photons range from 1 keV to 100 GeV, and the electrons range from 1 keV to 1 GeV [19]. Figure 1 shows the activation calculation procedure currently used in this study.

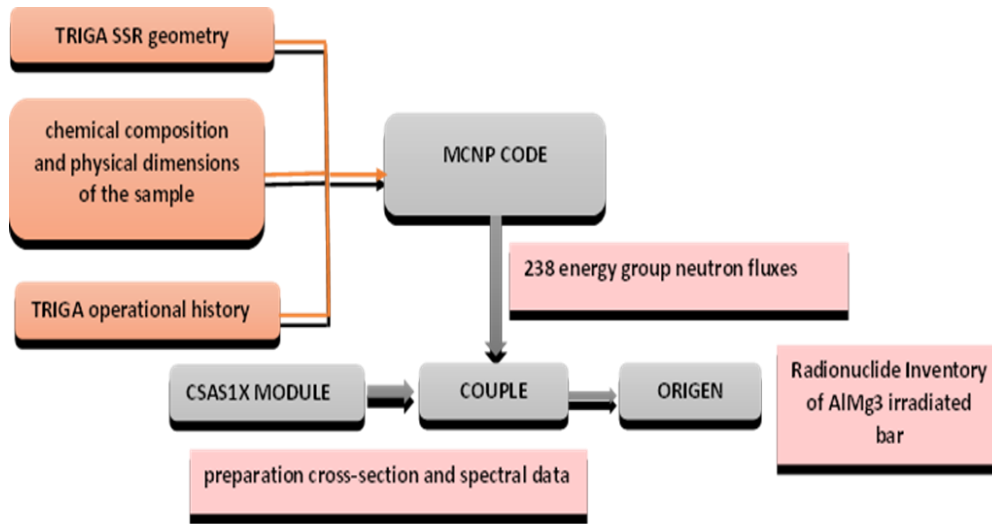


Fig.1. The graphical scheme used for activity inventory calculation

The neutron fluxes and the spectrum on the 238-energy group specific for the V7-238 standard SCALE multigroup cross-section library were calculated with Monte Carlo MCNP code [19] considering the geometry and chemical composition determined by ICP OES measurements of the irradiated sample.

The F4 tally in MCNP calculates the average flux in a cell or region by determining the total track length of particles within the cell, divided by its volume and normalized per source particle. This approach aligns with the physical definition of neutron flux:

$$\bar{\phi}_V = \frac{1}{V} \int dE \int dt \int dV \int d\Omega \Psi(\vec{r}, \Omega, E, t) \quad (1)$$

Where: $\bar{\phi}_V$ = average neutron flux in a cell (volume);
 V = volume (cm³);

E = energy (MeV);
 t = time (sh; 1sh = 10^{-8} s);
 Ω = direction vector;
 Ψ = angular flux (particles/cm²/sh/MeV/steradian);
 \vec{r} = particle position vector (cm).

The configuration of the TRIGA SSR 14 MW active zone and the AlMg₃ sample in G7 irradiation location modeling using MCNP code are presented in Figure 2.

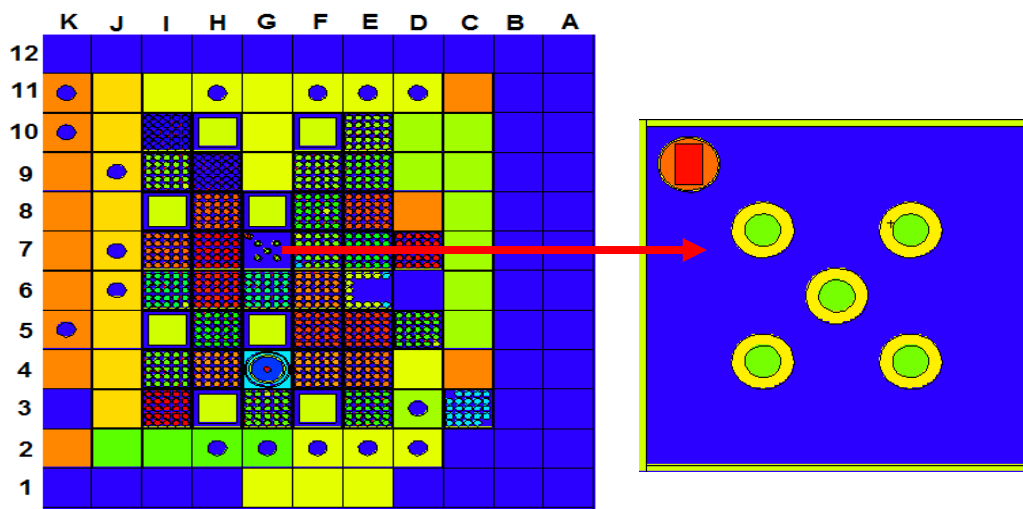


Fig.2. TRIGA SSR 14 MW and irradiation location (G7) of AlMg₃ sample MCNP model

The activation calculation procedure currently employed (see Fig. 1) uses the following SCALE modules: (1) CSAS1X for modeling the basic cell (homogenized fuel, AlMg₃, water, etc.) to replace the AlMg₃ infinite dilution cross-sections in the AMPX library; (2) COUPLE to generate a specific ORIGIN-S library for the neutron spectrum calculated with MCNP on 238 energy groups; (3) ORIGIN-S [20] to determine the activity for 1g of AlMg₃ assuming the homogeneity of the material.

The ORIGIN-S code uses the matrix exponential expansion model to calculate time-dependent concentrations, activities, and radiation sources for multiple isotopes that are produced or decay simultaneously by neutron conversion, fission, and radioactive decay. It has been developed and maintained as the depletion and decay module in the SCALE code system and can be used and executed as a standalone code using a wide range of cross-section libraries developed for different reactor types and fuel designs within ORIGIN-ARP in SCALE [20].

The procedures for generating the binary format problem-dependent ORIGEN-S library are included in the COUPLE code, which prepares a transition matrix containing the decay and cross-section transition rate constants. Multigroup cross-section data in the SCALE code system are obtainable via ENDF/B-V, -VI, and -VII files. The processing of these cross-sections takes into consideration the resonance self-shielding of the specific configuration that the user has set. The neutron flux spectrum obtained from the transport code analysis is then used to collapse them into a single group, which is then applied straight to the ORIGEN-S depletion calculation [20].

3. Results and discussions

3.1 SEM-EDS and EDXRF analyses

The results obtained by SEM-EDS and EDXRF analyses highlighted the non-homogeneity of the AlMg₃ analyzed sample. Also, metal inclusions were identified in the investigated sample. The main chemical elements identified were Al, Fe, Mn, Mg, Cr, Ni, Ti and Si. Figure 3 presents the elements distribution (mapping) in the investigated sample provided by SEM-EDS analyses.

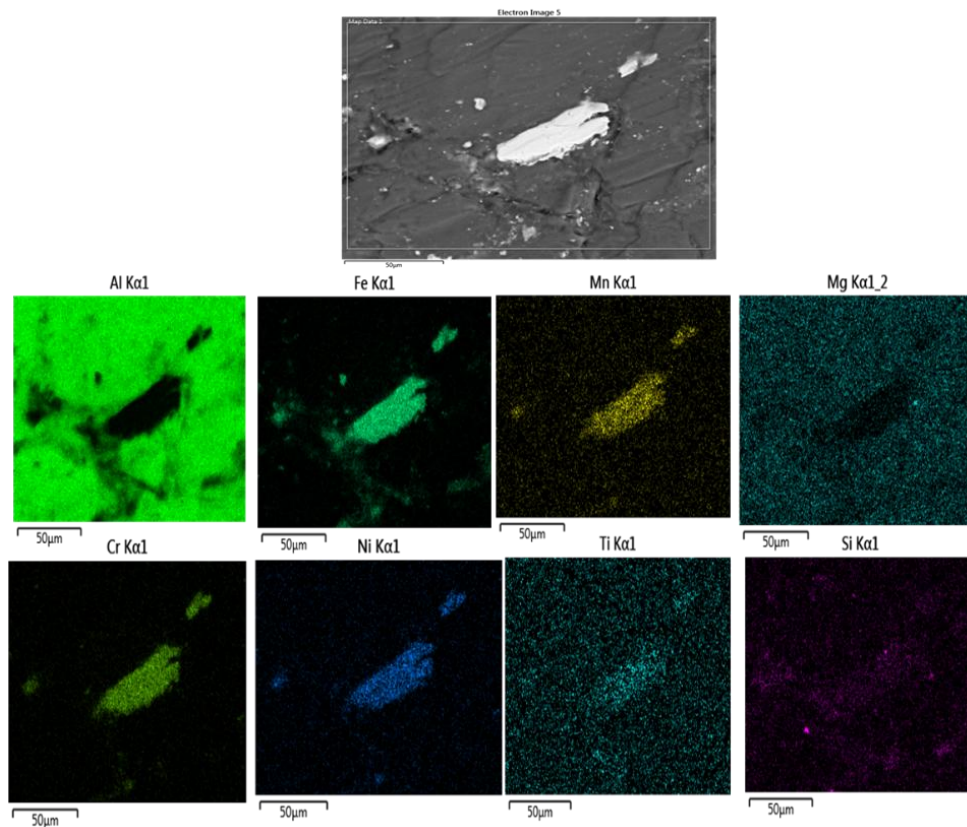


Fig. 3. The mapping distribution of the elements obtained by SEM-EDS

Additionally, EDXRF analyses were performed in three points of the AlMg₃ sample surface using the fundamental parameters method [21]. The concentration values obtained for the elements of interest through SEM-EDS and EDXRF are similar. Table 1 presents the average concentration values for each element obtained by both techniques.

Table 1

The average of the elements chemical concentration obtained with EDS and EDXRF techniques

Elements	Chemical Composition [wt%]	
Al	99.18±0.05	98±0.05
Si	0.25±0.03	0.18±0.003
Mn	0.07±0.03	<0.025
Zn	1.26±0.09	0.05±0.0003
Fe	0.47±0.03	0.35±0.001
Ti	0.4±0.07	0.008±0.002
Cr	0.41±0.08	<0.020
Co	-	<0.020
Ni	-	<0.015
Cu	-	<0.100
Techniques	SEM-EDS	EDXRF

Significant differences between the two sets of values were observed for the following elements: Mn, Zn, Ti, and Cr. These discrepancies can be attributed to both the volume and the inherent non-homogeneity of the investigated AlMg₃ alloy sample. The data obtained by these techniques are informative and provide information about the content of the main elements. The uncertainties associated on the results presented in table 1 arose from the uncertainties of the characterization techniques (SEM-EDS and EDXRF).

Considering the non-homogeneity of the AlMg₃ alloy sample and the fact that SEM-EDS and EDXRF techniques are relative surface techniques, which limit their accuracy, ICP-OES measurements were performed. Also, for the computer code modeling an accurate chemical composition of the irradiated material is required. Therefore, the ICP OES technique was used to quantify with adequate precision the amounts of impurities present in the analyzed material.

3.2. ICP OES measurements

ICP-OES provides accurate, quantitative data suitable for modeling code calculations. Its ability to detect elements at low concentrations improves our knowledge of the alloy composition.

The resulting data from ICP-OES measurements were analyzed and the data are graphically presented in Figure 4 as minimum, maximum, and average values.

To accommodate the lower concentration levels inherent in the material the concentration values of each element were presented on a logarithmic scale.

Given the small number of samples analyzed, we recommend expanding the dataset by investigating additional representative samples. Robust statistical conclusions need large sampling tests. The impurities identified by ICP-OES measurements in the AlMg₃ alloy include Li, Co, Sc, Ti, Mn, Cu, Cr, Mg, and Ni. The Al, Fe, Zn, and Si are the major elements identified by ICP-OES.

The average of impurities concentrations falls within the range of $(1.94 \times 10^{-1} \div 4.75 \times 10^{-5})$ mg/g as follows: $(4.21 \times 10^{-4} \pm 7.59 \times 10^{-6})$ mg/g Co, $(6.28 \times 10^{-3} \pm 5.12 \times 10^{-4})$ mg/g Li, $(4.75 \times 10^{-5} \pm 3.70 \times 10^{-6})$ mg/g Sc, $(2.81 \times 10^{-1} \pm 1.03 \times 10^{-2})$ mg/g Mn and $(1.94 \times 10^{-1} \pm 1.37 \times 10^{-2})$ mg/g Ti.

The uncertainties on the results arose from the following sources: uncertainties of mass and volumetric equipments (analytical balance and 50 mL class A volumetric flask) and uncertainties of spectral analysis method (ICP OES) [22].

The presence of Li and Co elements, even in small concentrations, can generate by neutron activation a high amount of radioactivity due to their Co and H isotopes. AlMg₃ alloy predominantly contains Al in high concentrations between 95÷99%, Mg, Mn, Si, and Fe in low concentrations, and in very low concentrations, elements such as Co, Cr, Li, Sc, Ti, Zn, and Ni.

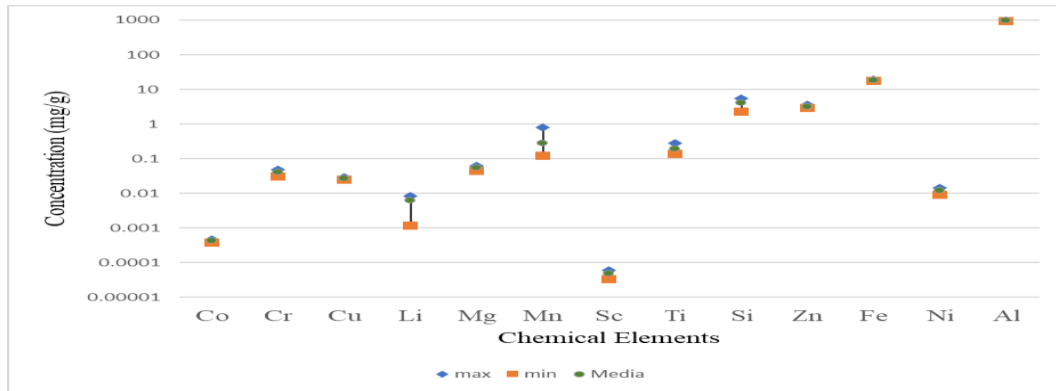


Fig.4. The chemical elements composition obtained by ICP-OES

The modeling input for the MCNP6 code was created using the average chemical composition values. This approach assumed the homogeneity of the AlMg₃ alloy sample. However, it is important to acknowledge that this assumption introduces additional uncertainties to the data obtained by MCNP6.

Antti Rätty et al in their studies related to the radiological characterization of various materials resulting from decommissioning of FiR1 TRIGA Mark II type research reactor, obtained for the irradiated fluental samples (a mixture of 30% metallic Al 69% AlF₃, and 1% LiF) around $10^5 \div 10^6$ Bq/g of the tritium

corresponding to a ^6Li average concentration of 149,174 ppb [6]. The fluental material served as a neutron moderator in the FiR1 TRIGA reactor and was studied due to the substantial neutron absorption cross-section of ^6Li which generates a significant amount of tritium.

In our study, we identified an impurity of approximately 6 ppm ($6 \times 10^{-3} \pm 5.12 \times 10^{-4}$ mg/g) of Li through ICP-OES measurements. Additionally, modeling computation codes estimated a tritium activity concentration of 10^6 Bq/g. In conclusion, we would like to underline the fact that Li presence in this alloy will generate via neutron activation a significant amount of tritium. Therefore, the assessment of Li impurity in the AlMg₃ alloy is an important activity to estimate the tritium activity. Given that tritium is a beta emitter with a half-life of 12.3 years, and an easily absorbed radionuclide, special attention needs to be given to the dismantling activities for the safe exposure of the working personnel.

3.3. Computer code results

The output file generated by the ORIGEN-S code provides detailed information on the activity concentration radionuclides within the irradiated AlMg₃ sample. The activity concentration of the radionuclides identified at the moment of discharge from the reactor and after and during the subsequent six months of cooling time is presented in Figure 5. A variety of short-lived radionuclides in high concentrations is observed at the discharge moment. Among these radionuclides we find ^3H , ^{55}Fe , ^{60}Co , ^{63}Ni , ^{54}Mn , ^{65}Zn , and ^{46}Sc representing the long-lived radionuclides.

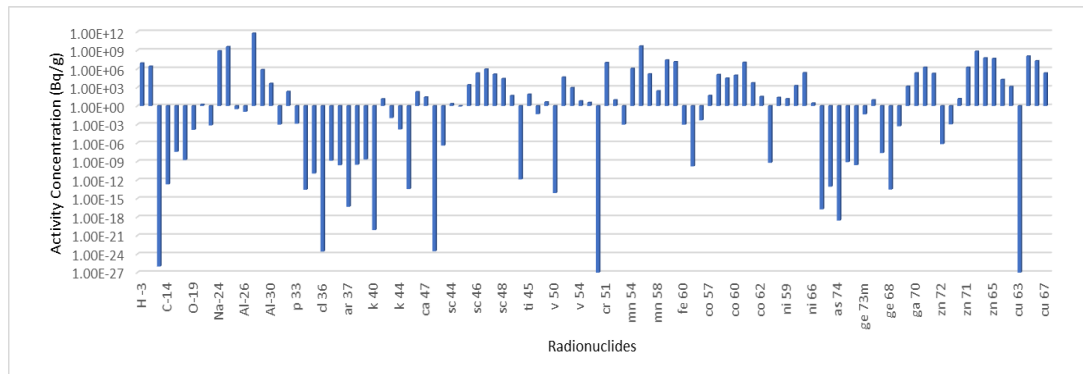


Fig. 5. The radionuclides inventory of the AlMg₃ alloy sample at discharge moment from the reactor

Figure 6 illustrates a comparative analysis of the activity concentration values obtained by the ORIGEN code for the long-lived radionuclides present in the irradiated AlMg₃ sample, at the discharge moment from the reactor and after 180 days of cooling time of the sample. The values following the six-month cooling time are used for future experimental tests as initial activity concentration.

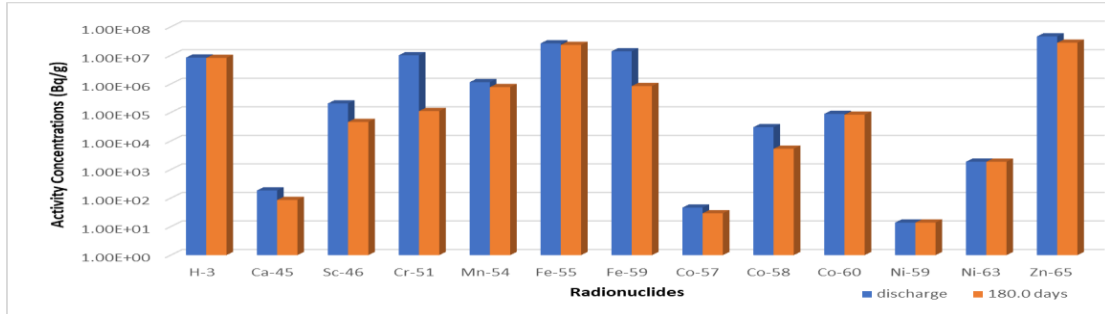


Fig. 6. The activity concentrations evolution of the long-lived radionuclides inventory at sample discharge from the reactor and after 180 days cooling time

Significant differences were observed in the activity concentrations of several radionuclides after 180 days, namely: ^{46}Sc , ^{51}Cr , ^{59}Fe , ^{45}Ca , and ^{58}Co radionuclides which have a decay time of up to 100 days. The activity values for ^{60}Co , ^{55}Fe , ^3H , and even for ^{65}Zn , ^{54}Mn radionuclides remained nearly constant during these 180 days of cooling.

Taking into consideration the experience of various countries engaged in the nuclear facilities decommissioning it is evident that the dismantling activities occur approximately a decade after facility shutdown. Considering the ORIGEN-S results, for the dismantling activities and safe management of the decommissioning waste, we will focus on those containing radionuclides with a half-life higher than a few hundred days.

Figures 7 and 8 illustrate the evolution of the activity inventory of long-lived radionuclides in the AlMg₃ alloy irradiated sample after 10 years of cooling time.

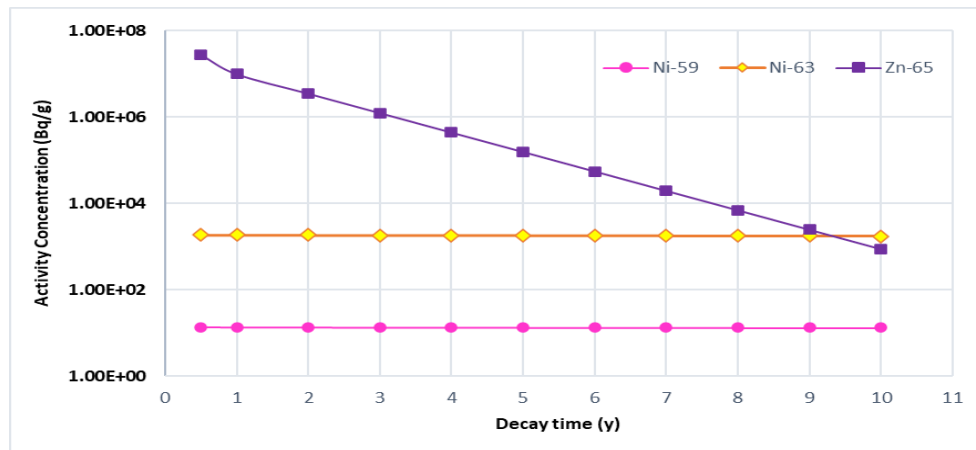


Fig. 8. The evolution of ^{59}Ni , ^{63}Ni and ^{65}Zn radionuclides after 10 years cooling time

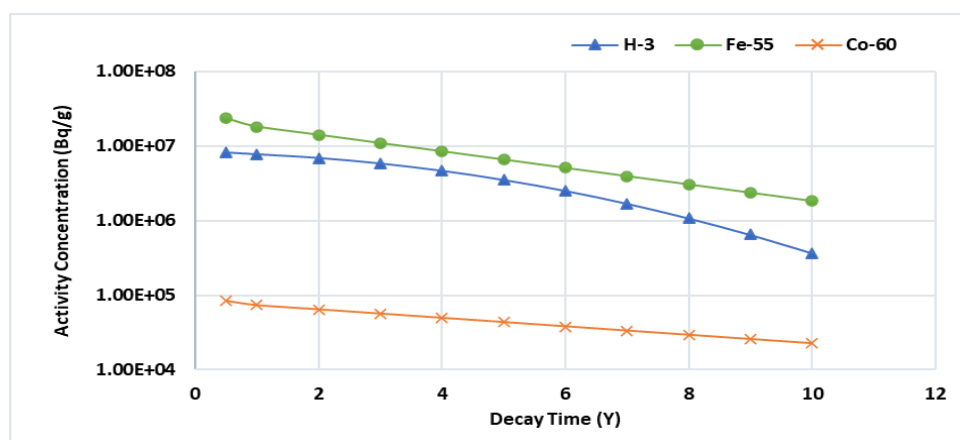


Fig. 7. The evolution of ^3H , ^{55}Fe and ^{60}Co radionuclides after 10 years cooling time

The main radionuclides with an important contribution to the total activity inventory of the irradiated aluminium after 10 years of cooling time are ^{55}Fe (82%), ^3H (17%), and ^{60}Co (1%). The ^{63}Ni and ^{65}Zn radionuclides are also present in small activity concentrations but their contribution represents less than 0.1 % of the total activity of the aluminium.

The activity concentration of ^{55}Fe and ^3H is relatively high, approximately 1.84×10^6 Bq/g and 3.7×10^5 Bq/g of aluminium. ^{55}Fe decays by electron capture, emitting X-ray while ^3H decays by beta emission; as both are pure beta emitters; they are considered difficult-to-measure radionuclides for which radiochemical separation methods are needed to be applied for their quantification. ^{63}Ni is also, a difficult-to-measure radionuclide with a half-life of 100.1 years, emitting pure beta particles for which the quantification will be challenging.

^{60}Co and ^{65}Zn are gamma emitters considered easy-to-measure by nondestructive methods such as gamma-ray spectrometry. The activity concentration of ^{60}Co and ^{65}Zn after 10 years of cooling of the irradiated sample will be around 2.2×10^4 Bq/g and 8.8×10^2 Bq/g, respectively.

Considering an estimated amount of 6 tons of aluminium waste resulted from TRIGA decommissioning, assuming a chemical composition similar to the AlMg3 alloy sample tested, the aluminium waste will have an activity concentration of approximately 2.2×10^6 Bq/g equivalent to 2.24 TBq/t or 13.4 TBq total radioactivity in the aluminium matrix after 10 years cooling time. The modeling codes that are based on the impurities content in the AlMg₃ alloy and the irradiation history can lead to overestimation of the inventory data.

Inventory data on TRIGA decommissioning waste are reported by Anti Rätty et al. [1, 6] and Quoc-Duong Tran et al. [4]. For around 2.2 kg of aluminium generated by the FiR1 TRIGA Mark II decommissioning, Rätty et al. determined a total activity of around 0.385 TBq (approximately 0.17 TBq/t). The main

radionuclides identified after six months from reactor shutdown were: ⁵⁵Fe (89%), ⁶⁵Zn (9.61%), (0.9%) ⁶³Ni, (0.42%) ⁵⁴Mn, (0.06%) ⁵⁹Fe, (0.01%) ⁶⁰Co and ⁴⁶Sc, ⁴⁵Ca, ⁵¹Cr (<0.01%) [1,6].

Quoc-Duong Tran et al. found in 129 kg of aluminium used in the reflector surrounding the DNRR (Dalat nuclear research reactor) reactor core a total activity of 0.016 TBq. Other components generated 541 kg of aluminium waste with a total activity of approximately 0.03TBq after 10 years of reactor shutdown, equivalent to data obtained by Quoc-Duong Tran et al. for aluminium waste corresponding to around 0.068 TBq/t [4].

The discrepancies between our estimated total activity and literature data may be generated by the uncertainties in the alloy composition and modeling code limitations. Usually, the calculation values obtained by modeling codes are overestimating the actual inventory. Ongoing experimental tests and refined simulations using MCNP6 and ORIGEN-S codes will validate these preliminary results.

4. Conclusions

Decommissioning projects involve a series of critical steps including planning, cost estimation, risk assessment, waste management, and site remediation. These steps are tightly linked to the radioactivity inventory, which significantly influences decision-making throughout the decommissioning process. Also, the radionuclides inventory is a requirement for the safe management of the radioactive waste resulting from TRIGA reactor decommissioning. Specifically, establishing the radionuclide inventory is essential for selecting the management route for the radioactive waste generated by the TRIGA reactor decommissioning.

The primary objective of our study was to estimate the quantity and types of radionuclides present in the irradiated AlMg₃ alloy sample. We achieved this through MCNP6 and ORIGEN-S modeling code calculations. The total radioactivity of reactor structural materials is largely attributed to trace impurities. Therefore, precise determination of the chemical composition of the AlMg₃ alloy was essential for accurate calculations using modeling codes. To assess impurities, we employed several techniques such as: ICP-OES which allowed us to quantify impurity content, while SEM-EDS and EDXRF provided insights into major elements. Among the impurities detected in the tested sample were lithium (Li), cobalt (Co), titanium (Ti), zinc (Zn), and scandium (Sc), all present in trace concentrations. The activation of Li, Fe, and Co impurities by neutron irradiation contributes significantly to radioactivity levels. After a cooling time of 180 days, we obtained by ORIGEN-S code calculation the main radionuclides and their activity concentration in the AlMg₃ alloy irradiated sample: 8.14 MBq/g of H-3, 85.7 Bq/g of Ca-45, 46.5 kBq/g of ⁴⁶Sc, 112 kBq/g of ⁵¹Cr, 773 kBq/g of ⁵⁴Mn, 23.3

MBq/g of ^{55}Fe , 841 kBq/g of ^{59}Fe , 29.3 Bq/g of ^{57}Co , 5.33 kBq/g of ^{58}Co , 83.6 kBq/g of ^{60}Co , 13.8 Bq/g of ^{59}Ni , 1.87 kBq/g of ^{63}Ni and 27.7 MBq/g of ^{65}Zn .

Assessing the radionuclides inventory for the dismantling activities typically performed a decade after reactor shutdown, represents an important input for the safe management of radioactive waste resulted from TRIGA reactor decommissioning. These radionuclides include ^{55}Fe whose activity represents around 82% of the total activity of aluminium waste, ^3H represents around 17%, ^{60}Co constitutes 1%, and ^{65}Zn and ^{63}Ni contribute to less than 0.1%.

Future studies will focus on the experimental tests carried out on the same irradiated aluminium sample and the uncertainties due to the modeling. The results obtained will be used as preliminary information for developing a characterization plan for the aluminium waste that will be generated by TRIGA decommissioning.

This study provides preliminary results which need to be improved and validated by experimental tests to accurately estimation of the radionuclides inventory contained in the aluminium resulted from TRIGA decommissioning activities.

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REFERENCES

- [1]. A. Rätty, „Activity Characterization Studies in Fir 1 Triga Research Reactor Decommissioning Project”, Academic Dissertation, University of Helsinki, Doctoral School In Natural Sciences Dissertation Series, 2020.
- [2]. *International Atomic Energy Agency*, „Radiological Characterization of Shut Down Nuclear Reactors for Decommissioning Purposes”, Technical Reports Series No. 389, Vienna, 1998.
- [3]. *General Atomic Company*, Safety Analysis Report Of The Triga Steady-State Research/Materials And Testing Reactor, February 1974, pp18-68.
- [4]. Q.D. Tran, K.C. Nguyen, B. V. Luong, T. N. Huynh, Q. H. Pham, D.H. Dang Vo, and N.D. Nguyen, “Determination of the Activity Inventory in the Structural Components of the Dalat Nuclear Research Reactor for Its Decommissioning Planning”, *Hindawi Science and Technology of Nuclear Installations* Volume 2022, Article ID 5174696, 10 pages <https://doi.org/10.1155/2022/5174696>.
- [5]. A. Rätty, M. T. Tyrkkö, P. Kotiluoto, and T. Kekki, „Validation and Optimization of Activity Estimates of the FiR 1 TRIGA Research Reactor Biological Shield Concrete”, *Nuclear Science and Engineering*, vol. 3. June 2022, American Nuclear Society DOI: <https://doi.org/10.1080/00295639.2021.2011671>.
- [6] A. Rätty, P. Kotiluoto, „FiR 1 TRIGA activity inventories for decommissioning planning”, PREDEC 2016, February 16-18, Conference, Lyon, France.

- [7] A. Rättyä, T. Lavonena, A. Leskinena, J. Likonena, C. Postolache, V. Fugaru, G. Bubueanu, C. Lungu, A. Bucsa, „Characterization measurements of fluental and graphite in FiR1 TRIGA research reactor decommissioning waste”, Nuclear Engineering and Design, July 2019, DOI: 10.1016/j.nucengdes.2019.110198.
- [8]. H. Beya., F. Rowayda. Mahmoud, Ahmed El Saghir, M. K. Shaat, and Alya Badawi, “Decommissioning Plan and Cost Estimation of TRICO-II Research Reactor”, Arab J. Nucl. Sci. Appl., Vol. 55, 3, 10-20 (2022), Web site: ajnsa.journals.ekb.eg.
- [9]. D. Gurau, R. Deju. D. Stanga, A. Zorliu, S. Ionel, M. Dragusin, “Evaluation of the concrete protection activity profile of the VVR-S RN from IFIN-HH”, conference paper, October 2016, <https://www.researchgate.net/publication/310775874>.
- [10]. E. Ionescu, D. Gurau, D. Stanga, O. G. DULIU, “Decommissioning of the VVR-S Research Reactor – Radiological Characterization of The Reactor Block”, Romanian Reports in Physics, Vol. 64, No. 2, P. 387–398, 2012
- [11]. C. M. Bucur, C. Ichim, I. Florea, C. Diaconescu, “Cementitious Materials Characterisation by Inductively Coupled Plasma Emission Spectroscopy”, Journal of Nuclear Research and Development, No.26, December, 48-54 Pp.
- [12]. M. Uemoto, M. Makino, Y. Ota, H. Sakaguchi, Y. Shimizu, and K. Sato, “Determination of Minor and Trace Metals in Aluminum and Aluminum Alloys by ICP-AES, Evaluation of the Uncertainty and Limit of Quantitation from Interlaboratory Testing”, Analytical Sciences, June 2018, Vol 34, pag. 719-724.
- [13] G. TyLER, J. Yvon, S.A.A. Horiba Group, “ICP-OES, ICP-MS, and AAS techniques Compared”, ICP Optical Emission Spectroscopy, Technical NOTE 05.
- [14]. P. R. Bairros da Silva, C. N. Makarab, A. P. Munarob, D. C. Schnitzlerb, A. D. Wastowskic and C. Poletto, “Comparison of the analytical performance of EDXRF and FAAS techniques in the determination of metal species concentrations using protocol 3050B (USEPA)” INTL.J. River Basin Management, 2016, <http://dx.doi.org/10.1080/15715124.2016.1203792>.
- [15]. L. R. e Silva, R. J.de Castro, J. A. Minski da Motta, C. E. Santos, P. R. Bairros da Silva, R. C. R. Bottini, D. C. Schnitzler, A. D. Wastowski, “Analysis of Average Concentrations of Fe, Al, and K in Natural Waters by EDXRF via Thermoconcentration Protocol”, ISSN 2178-0471 vol. 11 n.2 Dez. 2020 pág. 76-89.
- [16], A. V. Girao, G. Caputo and M. C. Ferro, “Application of Scanning Electron Microscopy Energy Dispersive X-Ray Spectroscopy (SEM-EDS)”, Comprehensive Analytical Chemistry, Vol. 75. <http://dx.doi.org/10.1016/bs.coac.2016.10.002>.
- [17] S. R. Babu, S. K. Michelic, “Overview of application of automated SEM/EDS measurements for inclusion characterization in steelmaking”, MetalMat., 2024; <https://doi.org/10.1002/metm.18>
- [18] ASM international handbook, “Properties and Selection: Nonferrous Alloy and special-Purpose Materials”, Volume 2 of the 10 Edition Metals Handbook, 1992.
- [19] X-5 Monte Carlo Team, Los Alamos National Laboratory, “MCNP A General Monte Carlo N-Particle Transport Code”, Version 5, April 24, 2003.
- [20]. Oak Ridge National Laboratory, “Origen-S: Depletion Module to Calculate Neutron Activation, Actinide Transmutation, Fission Product Generation, And Radiation Source Terms”, Oak Ridge, Tennessee, June 2011.

- [21]. *J. Kawai, K. Yamasaki, R. Tanaka*, “Fundamental Parameter Method in X-ray Fluorescence Analysis”, Encyclopedia of Analytical Chemistry, 2019.
- [22]. *J. R. Taylor*, “An Introduction to Error Analysis the Study of Uncertainties in Physical Measurements”, Second Edition, University Science Books, Sausalito, California, 1997.