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TEMPORAL EVOLUTION OF ACTIVITIES IN WASTES FROM MO-99 PRODUCTION

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ABSTRACT

Mo-99 is commonly produced by fission of U-235 because this is still the only process capable of generating the required amounts of Mo-99 with high specific activity. This method involves the dissolution of the irradiated U-235 target, followed by several stages of chemical separation, which generate wastes in all steps of the process, some with high levels of radiation, which irrevocably need to be managed. This paper presents a simulation of the evolution of the radioisotopic inventory that is expected to be generated in the planned production of Mo-99 in Brazil, as a contribution to designing the waste treatment plan. The code SCALE 6.0 was used to generate activity data using as input the geometry and composition of the target, the irradiation time, neutron flux and Mo-99 production schedule. The radionuclides of greatest interest for the waste management were identified. Shielding and cooling determined the criteria for initial storage; the transport limits are important for the step of treatment and packing; radiotoxicity of each radionuclide is considered in possible future scenarios of migration from final repository.

1. INTRODUCTION

Technetium-99m accounts for more than 80% of all diagnostic nuclear medicine procedures in the world and it is exclusively produced from the decay of its 66 h half-life parent Molybdenum-99[1]. There are two major methods for producing Mo-99, reactor neutron activation of Mo-98 and fission of U-235. According to IAEA[2]

> ñ it is technically difficult to prepare Mo-99 and this, combined with issues of radioactive waste products in the manufacture and nuclear regulatory requirements and security, has resulted in only a few suppliers of the product worldwide. Any disruption in the production schedule of one of these suppliers could have a global impact on the supply of generators and the clinical practice of Nuclear Medicine.ò

For this reason, there has been realized a need for new suppliers and methods to ensure security of supply. The safety of the plants, processes, waste management, transport and storage of radioactive material are issues to be resolved.

Storage of radioactive wastes means a way of maintaining them such that: isolation, environmental protection and monitoring are done according to safety standards; and actions involving treatment, conditioning and disposal are facilitated and available.

As each country has a specific demand of Mo-99 and its production must take into account the time to deliver from the radiopharmacy to the nuclear medicine services, a large production and high activities are required in a successful process. The conditions can be achieved irradiating targets containing both low-enriched uranium (LEU) or high-enriched uranium (HEU) in research reactors.

The current use of HEU is decreasing because of some constraints of the Nuclear Proliferation Treaty (NPT) and many research groups around the world continue showing the technical feasibility of converting current HEU processes to LEU[3], since it is necessary a larger amount of uranium to compensate the minor enrichment.

Therefore our work is based on LEU targets and focused on the general nuclide production in terms of waste management for the present production demand.

2. METHODOLOGY

During the irradiation of U-235 targets, a wide range of nuclides is formed. The OrigenArp code [4], a module of depletion analysis for SCALE®, performs detailed time-dependent isotopic generation and depletion for 1946 nuclides for fission in uranium targets and activation analysis. ORIGEN calculations are performed using cross-section libraries of ENDF/B nuclear data. The output is the inventory of radionuclides in the uranium target after irradiation

The initial inventory of radionuclides evolves according to the Bateman equations, which were programmed in a computer and used to define the radioisotopic inventory for varying times after target irradiation. Then the relevant nuclides were defined according to the criteria mentioned above.

2.1 Definition of input parameters

The first step is to define the target geometry and its composition; there were three possible targets: UAlx plate (dispersion fuel), Ni-clad plate and Ni-clad cylinder; each of them results in different nuclide composition. These geometries and compositions are the most used by the current suppliers. Claddings can vary depending on the available technology and the dissolution process of the target after the irradiation. For this paper, only UAlx was studied, seeking to confirm the reliability of our method by comparing the results with those presented in papers with similar data inputs.

The irradiation time was set up to seven days in order to facilitate comparison with known data about nuclide production in other institutes. Neutron flux and power were defined based on a hypothetical common reactor for the production of molybdenum. The dissolution of irradiated target and chemical separation will not be discussed in this paper.
The period of time in which the temporal evolution of the activities is studied is up to 10,000 years.

2.2 Getting the Radioisotopic Inventory

The OrigenArp code performs rapid and accurate point-irradiation and decay calculations using problem-dependent cross sections. An initial radioisotopic inventory was provided by the code with the label EOI (end of irradiation) and respective atomic masses, half-lifes and amount produced, expressed in grams.
The study was conducted aiming at getting every singular data from one Mo-99 production cycle, once it is part of a program that will involve one production per week and its implications according to the physical and chemical characteristics.

2.3 Bateman equations

In order to get a temporal evolution of activities one must consider all processes of production of each nuclide, like all modes of fission of uranium, decay from other nuclides, and neutron activation. It is also necessary to consider the branching of the decay chains, depending on the state of energy and probabilities. The solution of the Bateman equation are represented below [5]:

$$N_i(t) = \lambda_1\lambda_2\ldots\lambda_{K-1}N_0\sum_{j=0}^{K}\frac{e^{-\lambda_j t}}{\prod_{\substack{k=1\\ k\neq j}}^{i}(\lambda_k-\lambda_j)}$$

This solutions were programmed in an electronic spreadsheet and used to calculate the activities of every different decay chain in the target. The data provided by OrigenArp as EOI for the UAlx target was used as starting point to test the methodology applied in this paper.

2.4 Definition of relevant nuclides

Depending on the input parameters, different ranges of nuclides are produced in the process, but in any case, containing several hundreds isotopes. For the purpose of planning the management of the radioactive wastes, only some nuclides are relevant. The total inventory can be reduced to a small, more manageable shift, contenting just the isotopes with longer half-lives, higher radiotoxicity and higher activities.

High activities require proper shielding during transport, processing and storage of the wastes, in order to protect people from unnecessary radiation exposures. The decay heat of accumulated wastes may also require cooling of the storage tanks to limit the evaporation rate and to avoid the waste to boil.

Another significant issue that must be considered is the radiotoxicity. Estimating doses turns out to be extremely relevant because the risk of migration from final repository must be considered and is fundamental to safety assessment.

Most radioactive wastes must be stored prior to disposal, primarily because of technical considerations, for instance for decay of wastes containing mainly short lived radionuclides or for cooling of high level wastes. In other cases, storage may be practiced for reasons of economics or policy [6].

In a time frame of 10,000 years, adopted in the present work as the relevant horizon for assessment, just the relevant long-lived isotopes are of interest for waste management.

3. RESULTS

The graphs in figure 1 and figure 2 present the radionuclide inventory of the waste considering the previously mentioned criteria and the Brazilian regulations for classification of radioactive wastes [7].

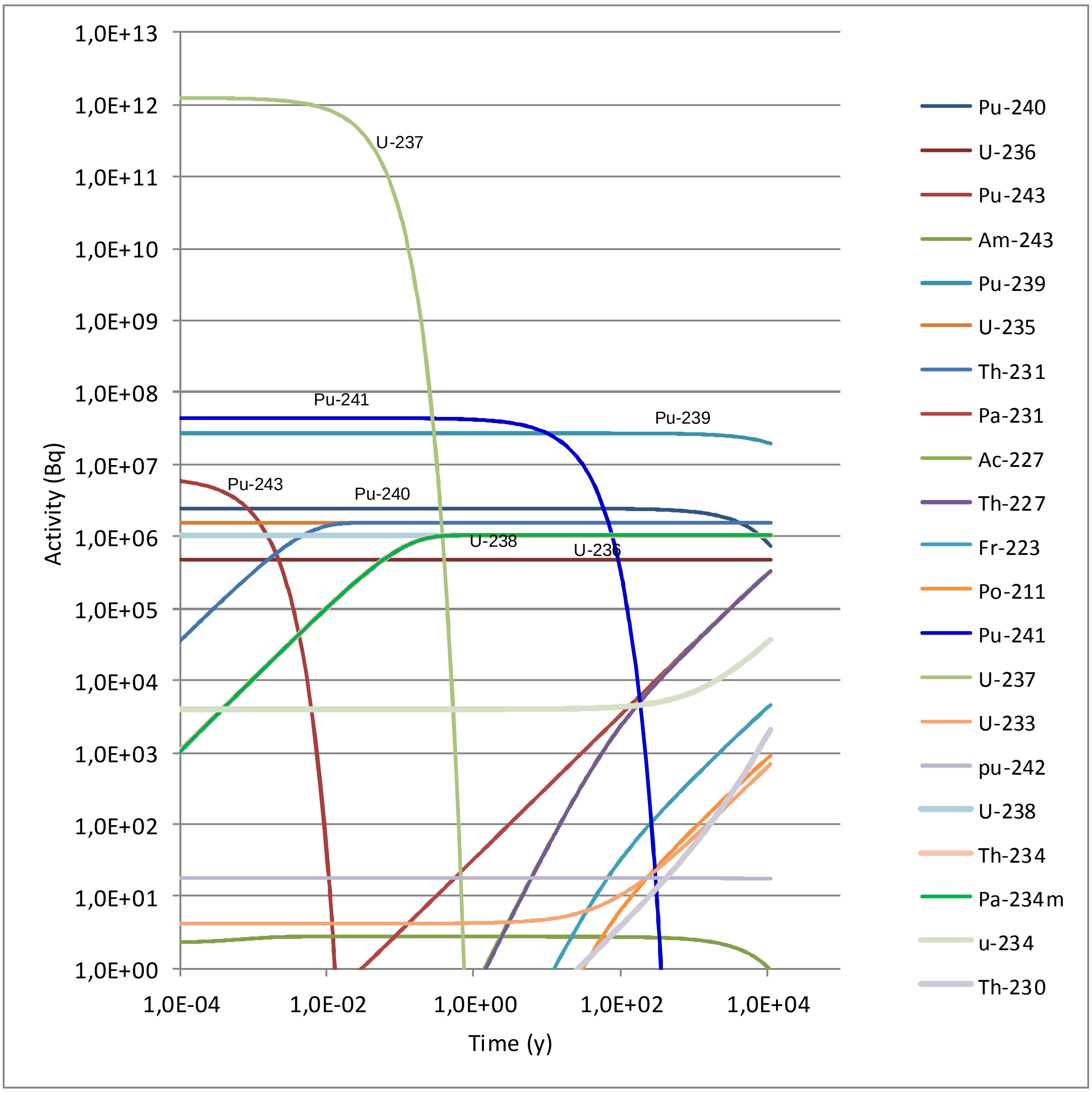

Figure 1. Temporal evolution of activities of long-lived nuclides.

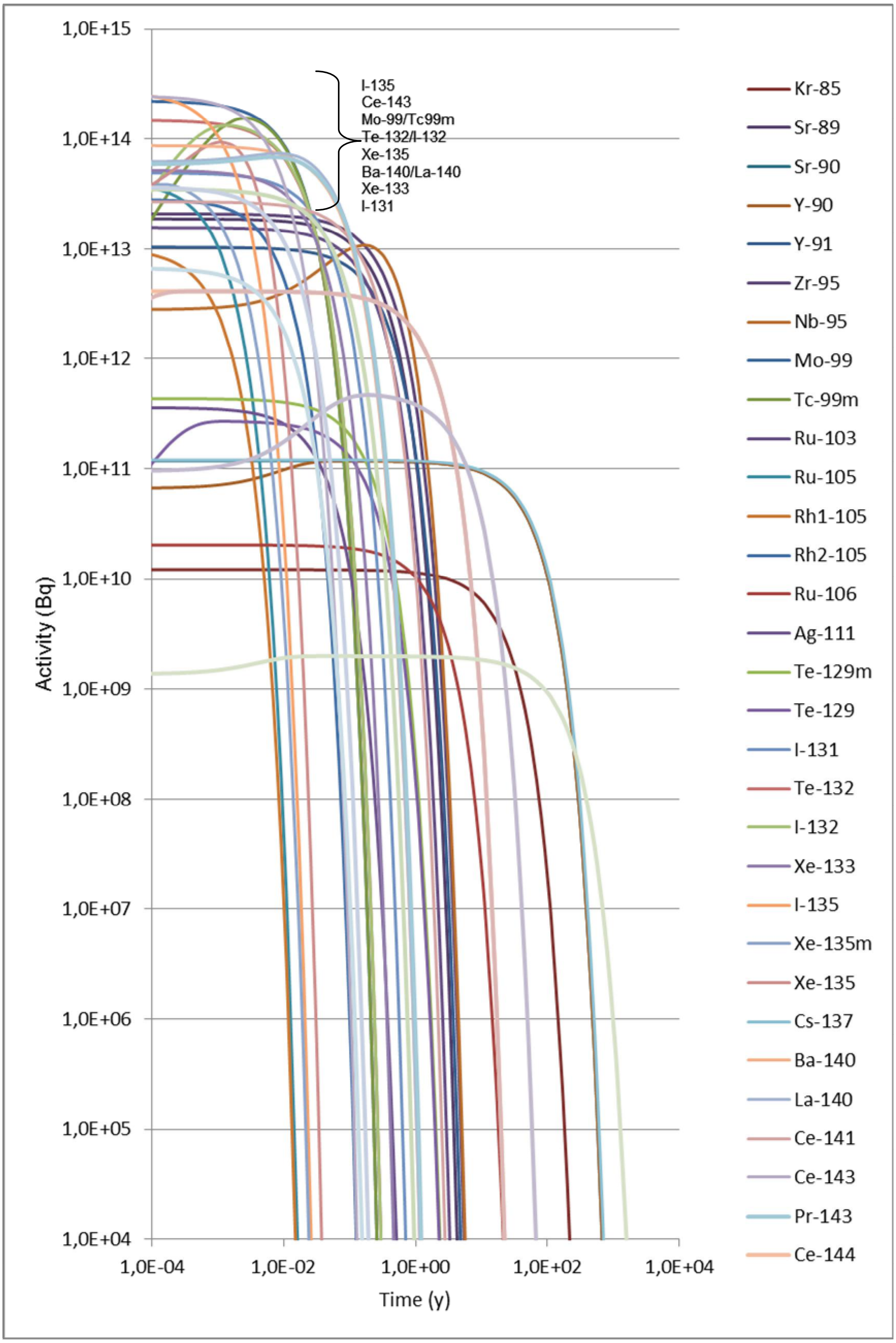

Figure 2. Temporal evolution of activities of short half-life nuclides.

Nuclear data used for decay heat calculation was obtained from IAEA Data Bases [8]. The next graphs (figure 3 and figure 4) present main values of decay heat based in activities shown previously.

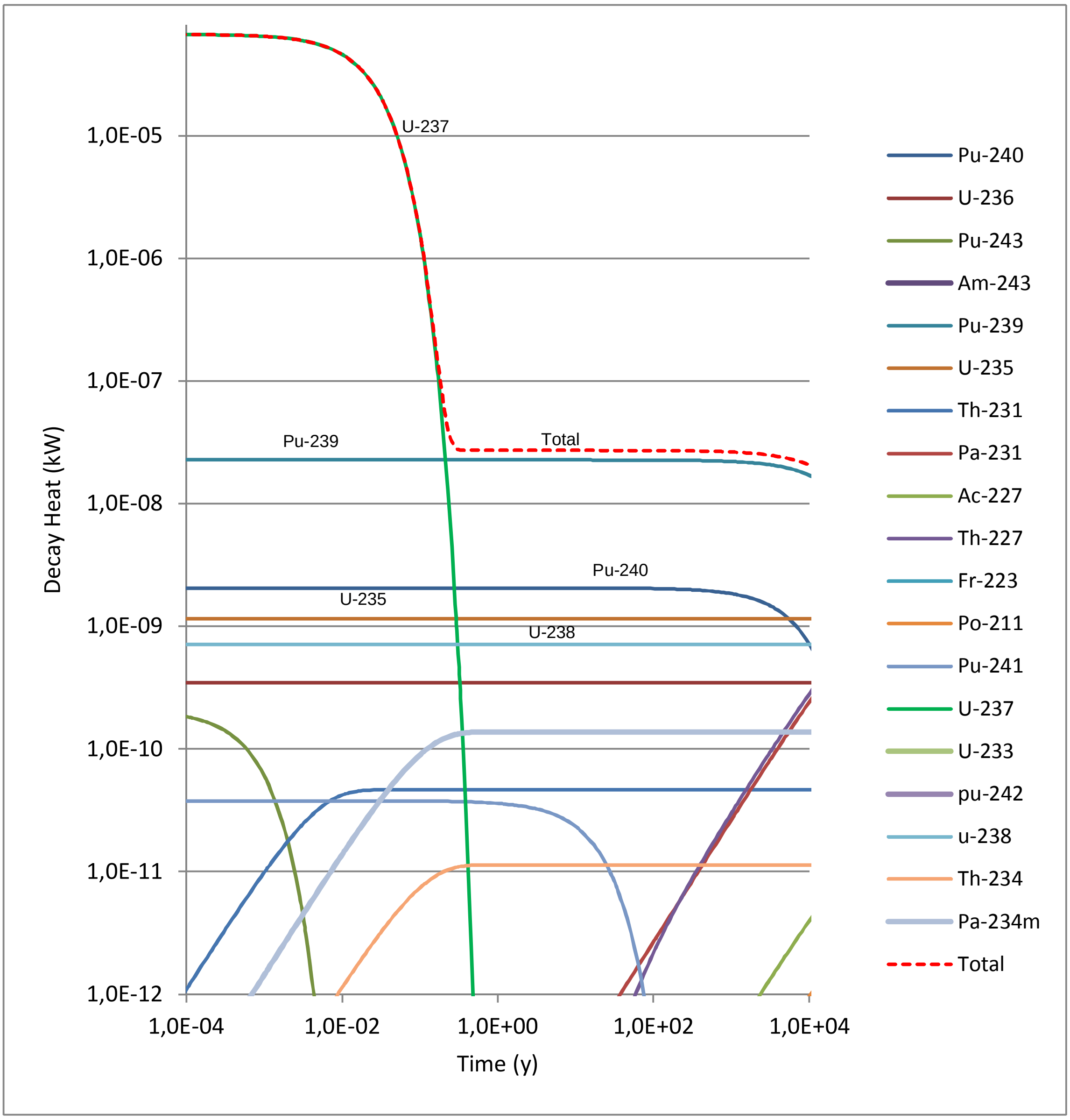

Figure 3. Decay heat of long-lived nuclides

The evolution of the radiotoxicity of the radioisotopic inventory over time was estimated using the committed effective dose potential of the inventory as indicator. This potential was calculated as the product of the activity of each radionuclide by the ingestion dose factor for adult individuals of the public.

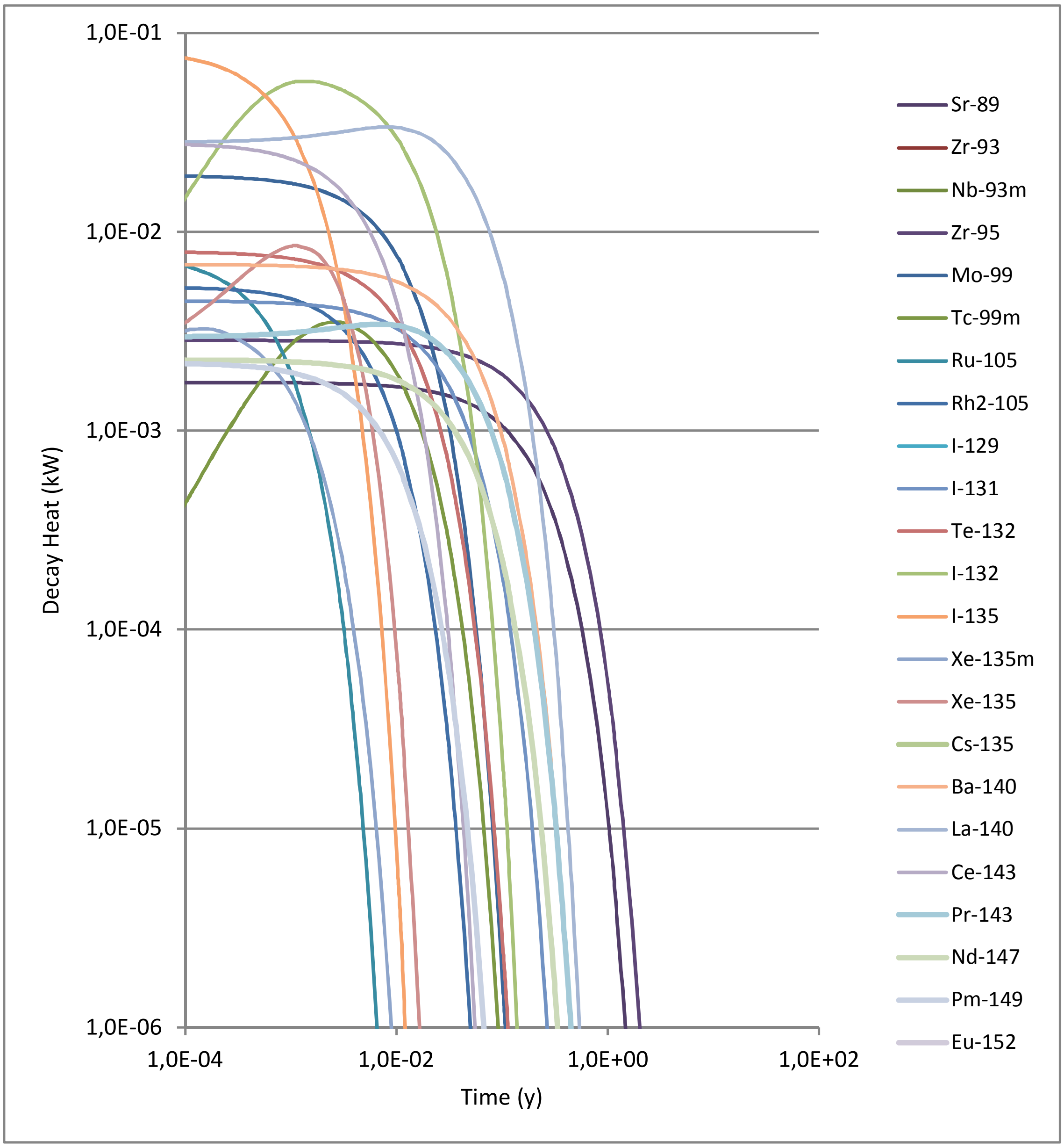

Figure 4. Decay heat of short-lived nuclides

Fig. 5 shows the remaining potential radiotoxicity until 300 ï 500 years, the range of time usually adopted to plan the release of the low- and intermediate-activity waste repositories from regulatory control, for unrestricted use. It can be seen that only few radioisotopes have potential to deliver doses above 0,1 mSv after one hundred years. These are Sr-90/Y-90, Zr-93, Tc-99, Sn-126, Sb-126, I-129, Cs-135, Cs-137, Sm-151.

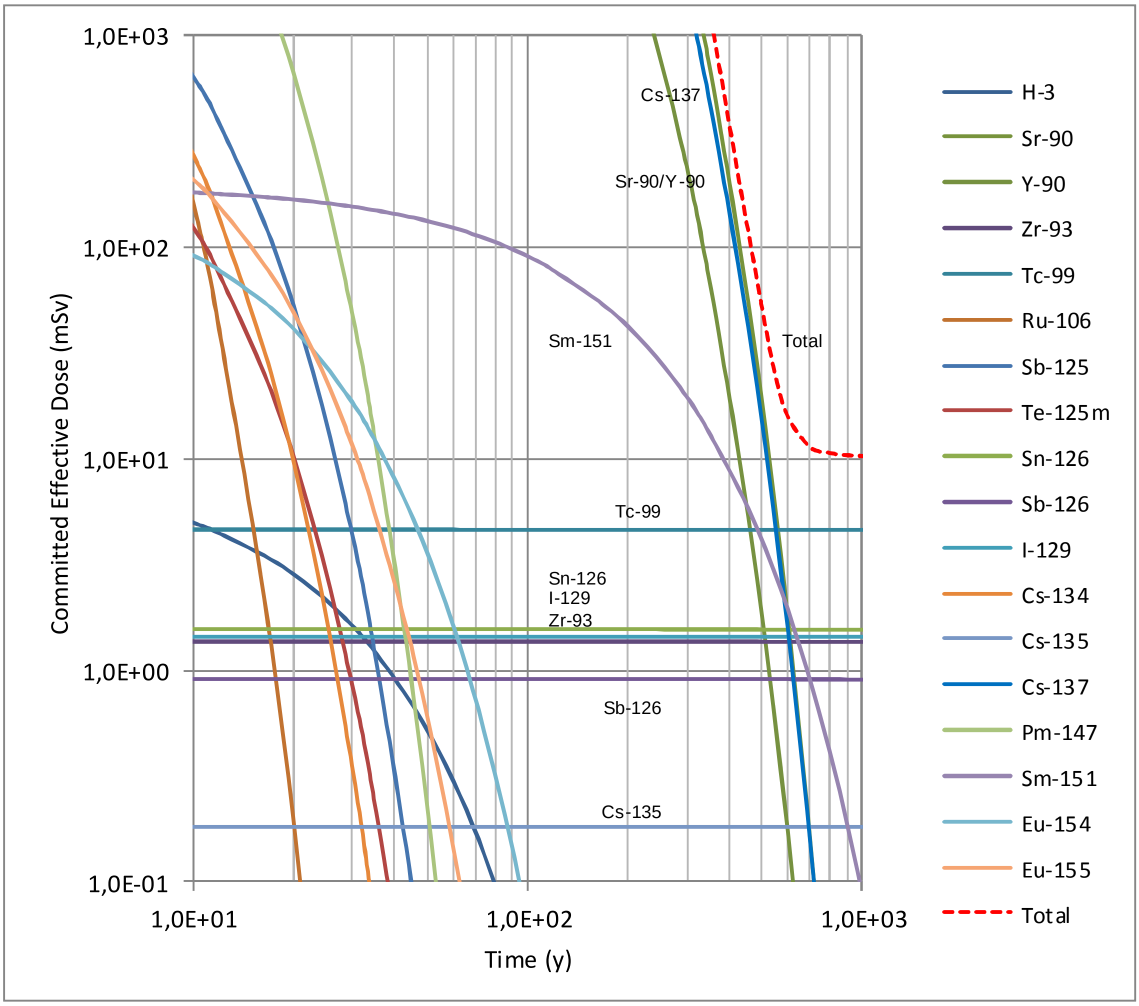

Figure 5. Potential radiotoxicity for some selected radioisotopes

A similar approach was used to determine the relevant radionuclides for external radiation, the thickness of lead necessary to reduce to 2 mSv/h the exposure rate at 1 m from a point source with the total activity of the radionuclide, being used as the indicator. The lead shielding thickness (x) was calculated with the simplified expression:

$$x = \frac{\ln\left(\frac{D1}{D0}\right)}{\ln(2)}.HVL$$

where
D1 = unshielded dose rate at 1 m from a point source;
D0 = shielded dose rate, arbitrarily set at 2 mSv/h, as a reference value;
HVL = half-value layer of lead for the radionuclide gamma energies and intensities;

Results are presented in Fig. 6, showing that the radionuclides Zr-95, Mo-99, Ru-103, I-132, and La-140 are relevant for only the first months after Mo-99 production, and that, for times later than about 1 year, only Cs-137 is relevant.

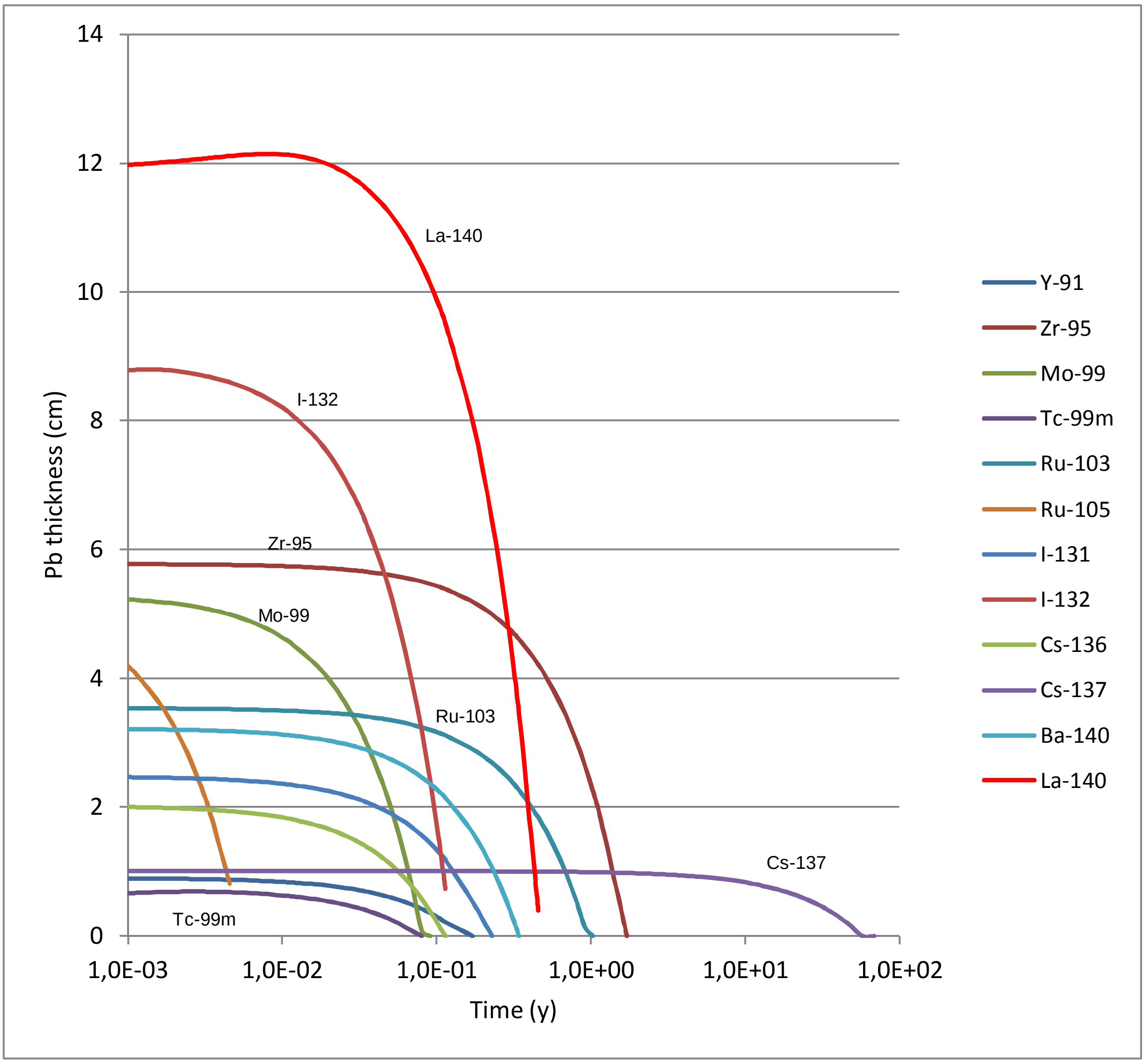

Figure 6. Estimation of shielding thickness requerid for internal transport

It is important to emphasize that these calculations are only deemed to produce preliminary indicators of radiotoxicity for final disposal and of relevance for shielding purposes, and that it must be kept in mind that the total activity of each radionuclide produced in one Mo-99 production campaign was used to calculate de dose rates. Actual dose rates will be calculated for the accumulated volumes of wastes from a series of production campaigns taking into account that the production process separates the radioisotopic inventory in many waste streams. Transuranium elements and their decay series will be dully considered in the ensuing work to further refine these results.

4. CONCLUSIONS

Results show that the decay heat of the wastes is not expected to exceed $2kW.m^{-3}$, suggesting that no high level radioactive waste will be generated in the Mo-99 production. However, the data presented in this paper must be further detailed, considering other target compositions and irradiation times. The research is in progress and is part of a larger scope project aiming at studying the management of the radioactive wastes generated in Mo-99 production facilities.

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