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RADIOACTIVE DECAY PATTERN OF ACTINIDES PRESENT IN WASTE FROM Mo-99 PRODUCTION

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ABSTRACT

Brazil is currently planning to produce ^{99}Mo from fission of LEU targets to meet the present national demand of ^{99m}Tc. The ^{99}Mo activity planned at the end of irradiation is 5000 Ci (185 TBq) per weekly cycle, in order to meet the present demand of 1000 Ci (37 TBq) per week, after target cooling and processing. To predict the activities that will be handled in the waste treatment facility, the computational code SCALE 6.0 was used to simulate the irradiation of the uranium targets and the decay of radioactive products. This study presents the findings of this research, mainly focused on the actinides activity that will be present in the waste and the respective radioactive decay pattern over a period of one hundred thousand years.

1. INTRODUCTION

The ^{99m}Tc is one of the most worldwide radioisotopes used in nuclear medicine applications. Currently, the entire national demand is met by importing ^{99}Mo from Australia, Argentina and Russia. In order to supply the national market, Brazil is currently planning to produce ^{99}Mo from fission of low-enriched uranium targets, in an enterprise associated with the construction of the Brazilian Multipurpose Reactor (RMB).

To meet the present demand of 1000 Ci (37 TBq) to be weekly delivered to nuclear medicine services elsewhere in the country, the planned end of irradiation activity of ^{99}Mo is 5000 Ci (185 TBq) per weekly cycle of production.

The number of waste streams that will be generated in the plant and the characteristics of each stream depend on the process of U target dissolution and ^{99}Mo separation that will be adopted. However, the total activity of the actinides, fission and activation products that will be present in the waste can be predicted based on the yields of fission and activation process, associated with irradiation conditions, such as composition and mass of U targets, irradiation time, neutron flux, production schedule etc., which were, in principle, already established by the project management.

Radioactive decay of the waste over a very long period of time cannot be calculated by a single exponential, either classical Bateman equation will not correctly predict ingrown of daughters produced in a decay chain, unless activities just at the end of the Uranium target irradiation is known. This is more noticeable for actinides, due to the long chain of actinium, neptunium, thorium and uranium decay series.
The aim of this paper is to show the decay pattern for actinides present in the waste and to identify the most critical radionuclides in terms of activity over a period of one hundred thousand years.

2. METHODOLOGY

This study assumed that the U targets for ^{99}Mo production is composed by a 19.9% ^{235}U enriched UAl_x alloy dispersed in an aluminum Al-1050 matrix. The isotopic composition of the target is shown in Table 1; the elements magnesium, silicon, titanium, vanadium, manganese, iron, copper and zinc are impurities present in the Al-1050 alloy.

Table 1: Uranium target isotopic composition

Element	Mass (g)	Element	Mass (g)	Element	Mass (g)
^{24}Mg	$1.032\ 10^{-2}$	^{49}Ti	$4.200\ 10^{-4}$	^{65}Cu	$4.030\ 10^{-3}$
^{25}Mg	$1.310\ 10^{-3}$	^{50}Ti	$4.100\ 10^{-4}$	^{64}Zn	$6.310\ 10^{-3}$
^{26}Mg	$1.440\ 10^{-3}$	^{50}V	$3.000\ 10^{-5}$	^{66}Zn	$3.660\ 10^{-3}$
^{27}Al	$2.614\ 10^{1}$	^{51}V	$1.304\ 10^{-2}$	^{67}Zn	$5.400\ 10^{-4}$
^{28}Si	$6.027\ 10^{-2}$	^{55}Mn	$1.307\ 10^{-2}$	^{68}Zn	$2.490\ 10^{-3}$
^{29}Si	$3.060\ 10^{-3}$	^{54}Fe	$6.110\ 10^{-3}$	^{70}Zn	$8.000\ 10^{-5}$
^{30}Si	$2{,}020\ 10^{-3}$	^{56}Fe	$9.594\ 10^{-2}$	^{238}U	9.988
^{46}Ti	$6.500\ 10^{-4}$	^{57}Fe	$2.220\ 10^{-3}$	^{235}U	2.5
^{47}Ti	$5.800\ 10^{-4}$	^{58}Fe	$2.900\ 10^{-4}$	^{234}U	$7.500\ 10^{-2}$
^{48}Ti	$5.780\ 10^{-3}$	^{63}Cu	$9.040\ 10^{-3}$		

The irradiation time, the number of ^{235}U target plates and the reactor thermal neutron flux were chosen in order to meet the planned weekly demand of ^{99}Mo. Therefore, the values were set to seven days of continuous irradiation, 16 plates of ^{235}U targets and a thermal neutron flux of $1x10^4$ n.cm^{-2}.s^{-1}[1].

During the target irradiation, hundreds of nuclides are generated due to ^{235}U fission, activation products and radioactive decay chains. The OriginArp, a module of depletion analysis of the SCALE 6.0 code [2] was used to generate an output table with radionuclides activity over a period of 100.000 years.

3. RESULTS

Results from running the OrigenArp code showed that, keeping the above input specifications, 0.17 PBq of ^{99}Mo will be generated per weekly campaign, at the end of irradiation. The total

activity present in the U target is 6.03 PBq, comprising 5.65 PBq of fission products, 0.14 PBq of activation products and 0.24 PBq of actinides. Regardless in which waste stream the radionuclides will be present, the total waste radioactive decay over a 100.000 years period is shown in Fig. 1.

Fig. 2 and 3 show the decay pattern of the main actinides, arranged according to the actinium, neptunium, uranium and thorium decay series. Output from OrigenArp shows the activity of the one hundred of actinides present in the waste. Only those presenting activity higher than 0.1% of the total actinide activity in any year of the simulation are shown in the Figures.

In these Figures, radionuclides labelled * include the activity of the decay chain short-lived daughters. In the actinium series, ^{235}U* activity is the sum of the ^{235}U and ^{231}Th activities; for ^{227}Ac*: ^{227}Ac, ^{227}Th, ^{223}Ra, ^{219}Rn, ^{215}Po, ^{211}Bi, ^{211}Pb and ^{207}Tl. In the uranium series, for ^{238}U*: ^{238}U, ^{234}Th and ^{234}Pa; for ^{226}Ra: ^{222}Rn, ^{218}Po, ^{214}Pb, ^{214}Bi, ^{214}Po, ^{210}Pb, ^{210}Bi and ^{210}Po. In the neptunium series, for ^{237}Np*: ^{237}Np and ^{233}Pa; for ^{229}Th*: ^{229}Th, ^{225}Ra, ^{225}Ac, ^{221}Fr, ^{217}At, ^{213}Bi, ^{209}Tl, ^{213}Po and ^{209}Pb.

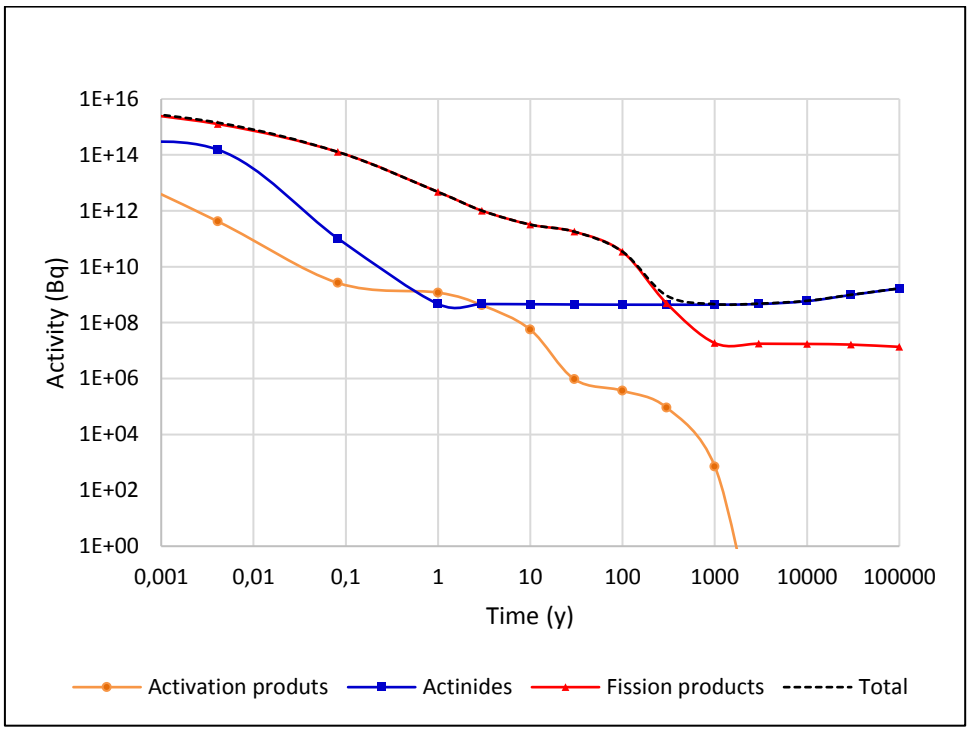

Figure 1: Radioactive decay of the waste from each weekly campaign of Mo-99 production.

Fig. 4 shows the activity percentage contributions of each radionuclide in relation to the total waste activity.

It can be noted that after approximately 300-500 years, the actinides – mainly ^{234}U, ^{239}Pu and ^{226}Ra plus short-lived daughters – account for almost all of the activity remaining in the waste. As expected, at the first 100-300 years, long-lived actinides have a small contribution to the

total activity; at this period of time, the majority of the activity in the waste comes from fission products $^{90}Sr/^{90}Y$, $^{137}Cs/^{137m}Ba$ and $^{144}Ce/^{144}Pr$.

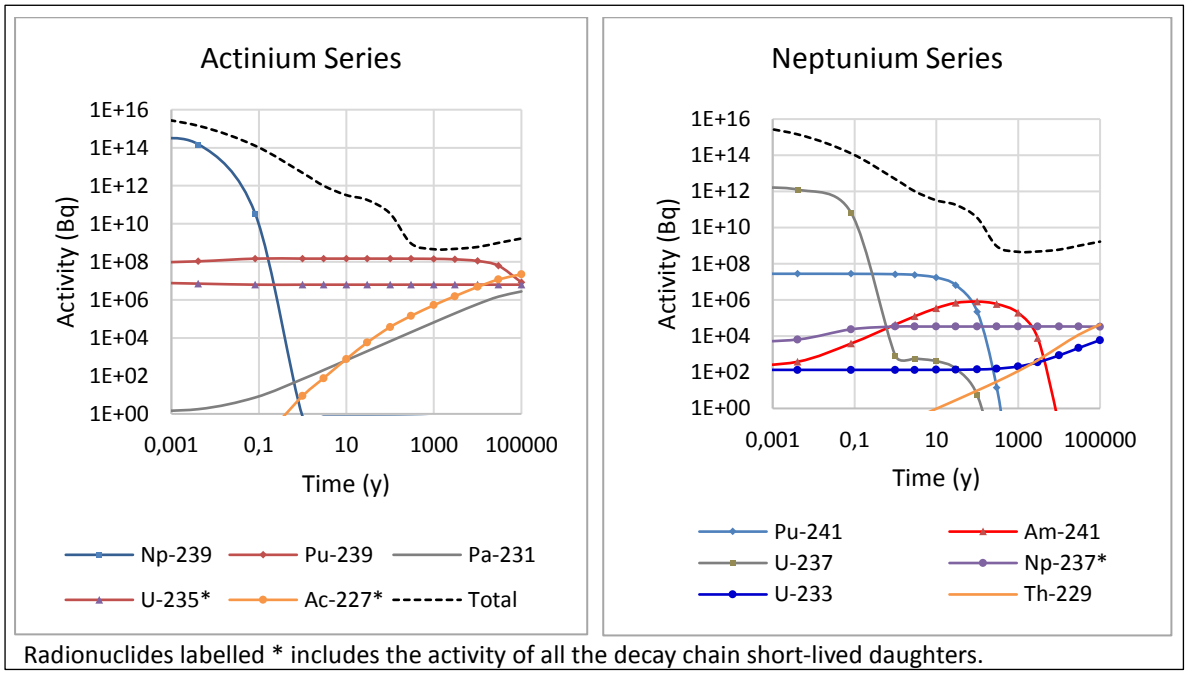

Figure 2: Radioactive decay of the radionuclides from actinium and neptunium series.

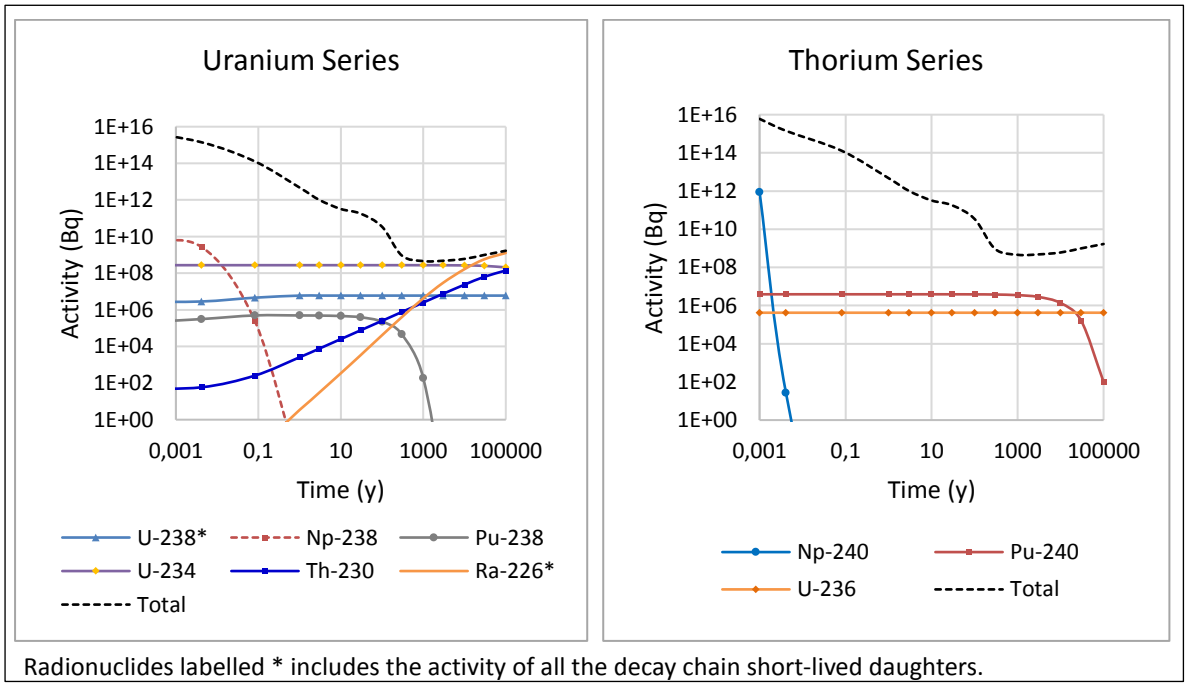

Figure 3: Radioactive decay of the radionuclides from uranium and thorium series.

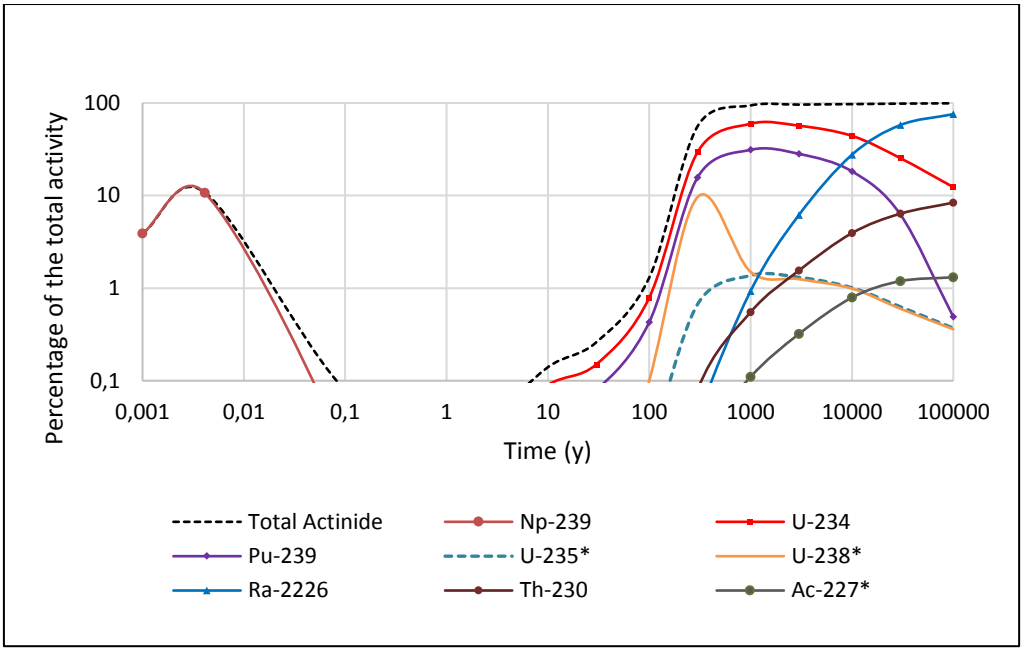

Figure 4: Percentage contributions of the main actinides to the total waste activity.

3. CONCLUSIONS

Although the results presented refer to the activities present in the waste generated in one week cycle of U target irradiation, results from this study could be easily extended to the cumulative waste resulted from various cycles of ^{99}Mo production.

An interesting point to be empathized is that the expected radioactive waste composition from a ^{99}Mo production plant is quite different from the waste arising from a PWR reactor type [3], in terms of dominant radionuclides activity. It is mainly due to – among others - the much shorter irradiation time of the U target, higher ^{235}U enrichment and different materials used in target and fuel elements.

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