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SUBSIDES FOR OPTIMIZATION OF TRANSFER OF RADIOACTIVE LIQUID WASTE FROM ^{99}MO PRODUCTION PLANT TO THE WASTE TREATMENT FACILITY

Maria Eugênia de Melo Rêgo, Roberto Vicente and Goro Hiromoto

Instituto de Pesquisas Energéticas e Nucleares (IPEN/CNEN – SP)
Av. Prof. Lineu Prestes, 2242
05508-000 São Paulo, SP
maria.eugenia@ipen.br
rvicente@ipen.br
hiromoto@ipen.br

ABSTRACT

The increasing need for radioisotopes lead Brazil to consider the domestic production of ^{99}Mo from fission of low enriched uranium targets. In order to meet the present demand of ^{99m}Tc generators the planned 'end of irradiation' activity of ^{99}Mo is about 170 TBq per week. The radioactive waste from the production plant will be transferred to a waste treatment facility at the same site. The total activity of the actinides, fission and activation products present in the waste were predicted based on the fission yield and activation data for the irradiation conditions, such as composition and mass of uranium targets, irradiation time, neutron flux, production process and schedule, already established by the project management. The transfer of the waste from the production plant to the treatment facility will be done by means of special shielded packages. In the present study, the commercially available code Scale 6.0 was used to simulate the irradiation of the targets and the decay of radioactive products, assuming that an alkaline dissolution process would be performed on the targets before the removal and purification of ^{99}Mo. The assessment of the shielding required for the packages containing liquid waste was done using MicroShield 9 code. The results presented here are part of a project that aims at contributing to the design of the waste management system for the ^{99}Mo production facility.

1. INTRODUCTION

Nowadays, Technetium-99m is the most important diagnostic radioisotope in nuclear medicine and it has been largely used in about 70% of all nuclear medicine procedures [1]. It is one of ^{99}Mo daughters, an artificial radioisotope mainly produced through uranium fission reactions in nuclear reactors [2].

After a worldwide shortage in 2009, caused by the interruption of operation of one of the world's major ^{99}Mo suppliers [3], many countries got involved in global efforts to ensure a reliable supply and reviewed potential methods of producing ^{99}Mo, of which the two main methods are reactor-based and accelerator-based [4].

Brazil has decided to produce ^{99}Mo from fission of low enriched uranium LEU targets [5] aiming at meeting safeguard requirements of the Non-Proliferation of Nuclear Weapons Treaty (NPT) [6]. Generation of high- and intermediate-level wastes is expected at the Brazilian ^{99}Mo processing plant. Proper management of these wastes is required in order to comply with national and international standards.

This project aimed at predicting the amount of liquid waste that will be generated at that facility and at performing the calculations on required shielding for interim storage and later transportation to the radioactive waste treatment plant.

The end of irradiation (EOI) activity of ^{99}Mo planned for this project is about 170 TBq per week in order to meet the present domestic demand of ^{99m}Tc generators.

2. METHOD

2.1. Prediction of the radionuclide inventory

The Origen S software [7], part of the Scale® code, was used to simulate the irradiation of targets. These targets are made of uranium-aluminum alloy, containing 2.5 g of ^{235}U, enriched to 19.9% and 20.6 g in total of 1050-aluminum alloy cladding per plate. Considering the 200 TBq of ^{99}Mo9 as a reasonable desired production per irradiation batch, this study considered sixteen (16) of these plates being irradiated simultaneously.

Origen S calculated the resulting radioisotope inventory of the targets considering a thermal neutron flux of $1x10^{14}$ $n.cm^{-2}.s^{-1}$ and an irradiation time of 7 days. The production scenario chosen for this paper was alkaline dissolution of the targets, followed by filtration and purification with ion exchange resins.

These settings resulted in a 169 TBq production of ^{99}Mo at the end of irradiation, according to the simulation. After a 12 hour cooling, as it is commonly used at other similar facilities, the total amount of ^{99}Mo would be 149 TBq, approximately 9 TBq per plate.

The radioisotope inventory was calculated for different periods simulating 3 months of production. The activities were summed up to represent the cumulative process that would happen inside the waste collecting tanks.

2.2. Liquid Waste Accretion

It was assumed that the alkaline dissolution results in the precipitation of 60% of the fission products generated during irradiation and the radioactive gaseous waste are taken to an *off-gas* treatment system. Therefore, gaseous and solid waste will be studied separately and only the results for liquid waste are presented in this paper.

The reference ^{99}Mo separation process [8] leads to two main streams of radioactive liquid waste: one acid stream resulting from the acidification of the process solution with HNO_3 for iodine removal and one alkaline stream resulting from the elution of ^{99}Mo from a separation column with NH_4OH. A chemical separation was simulated by retaining some radioisotopes in each waste flow, according to the dissolution and purification processes chosen for this study.

Each liquid wastes flow will be collected in separate cylindrical tanks dimensioned according to the amount of waste to be stored and with diameter to height ratios as to optimize the dimensions of cylinders.

It was assumed that these tanks would be located bellow the production hot cells and will remain there until the later transportation to the treatment unit *in-site*. They will collect the liquid wastes to their nominal capacity and will be replaced by a spare tank. After replacement the waste tank is sent to the treatment facility.

This study considered that each processing to separate iodine and molybdenum products, 30L of acid waste and 15L of alkaline waste will be generated. The collecting time ranges from 1 up to 12 weeks of production, so the volume, activity and shielding vary for each period considered.

2.3. Acquisition of shielding data

MicroShield® [9] is a comprehensive photon/gamma ray shielding and dose assessment program used for designing the shielding. Cylindrical geometry was used to represent the waste tanks and the dose rates were calculated using activities from Origen S calculation.

For 12 weeks of production, shielding with thickness varying from 1 cm up to 30 cm of lead was simulated in order to optimize the thickness of lead associated to the amount of waste and doses delivered, once the volume and activity increases as a function of time and the waste is cumulated in the shielded tanks.

3. RESULTS

Tables 1 and 2 show the results obtained for main radionuclides activity concentration in the acid liquid waste, considering one and three batches production per week, respectively. Calculated radionuclides concentration in the alkaline liquid waste are about one order of magnitude lower than in the acid waste.

All the radionuclides listed are fission products. For short time frame consideration and for shielding calculation purposes, the activity concentrations of the actinides in the liquid waste are negligible, since it was assumed that they all precipitate at the early phases of the target dissolution. Results for shielding calculations are presented for both, acid and alkaline liquid waste, considering one batch and three batches production per week.

Fig.1 shows the maximum dose rate delivered at the surface of the cylindrical shield, in function of different thickness of lead and time of the acid liquid waste accumulation, considering one batch production per week. The same is presented in the Fig.2, for alkaline waste.

Fig.3 shows the maximum dose rate delivered at the surface of the cylindrical shield, in function of different thickness of lead and time of the acid liquid waste accumulation, considering three batches production per week. The same is presented in the Fig.4, for alkaline waste.

It can be observed that, no matter the week or the waste stream considered, increasing ^{99}Mo production to three batches per week will not substantially affect the thickness of the shielding required.

Table 1. Acid waste in a one-batch per week production

Nuclide	1 week (Bq/mL)	3 weeks (Bq/mL)	6 weeks (Bq/mL)	9 weeks (Bq/mL)	12 weeks (Bq/mL)
Ag-111	$1{,}3x10^7$	$7{,}5x10^6$	$4{,}3x10^6$	$2{,}9x10^6$	$2{,}2x10^6$
Ba-137m	$8{,}5x10^6$	$8{,}5x10^6$	$8{,}5x10^6$	$8{,}5x10^6$	$8{,}4x10^5$
Cs-136	$3{,}8x10^6$	$2{,}8x10^6$	$1{,}8x10^6$	$1{,}3x10^6$	$1{,}0x10^6$
Cs-137	$8{,}9x10^6$	$9{,}0x10^6$	$9{,}0x10^6$	$8{,}9x10^6$	$8{,}9x10^5$
I-131	$1{,}1x10^8$	$6{,}5x10^7$	$3{,}8x10^7$	$2{,}6x10^7$	$1{,}9x10^7$
I-132	$9{,}1x10^7$	$3{,}9x10^7$	$2{,}0x10^7$	$1{,}3x10^7$	$9{,}8x10^6$
I-133	$1{,}2x10^6$	$4{,}1x10^5$	$2{,}1x10^5$	$1{,}4x10^5$	$1{,}0x10^5$
Mo-99	$2{,}9x10^8$	$1{,}2x10^8$	$5{,}8x10^7$	$3{,}9x10^7$	$2{,}9x10^7$
Nb-95	$6{,}7x10^7$	$9{,}4x10^7$	$1{,}2x10^8$	$1{,}3x10^8$	$1{,}4x10^8$
Nb-95m	$3{,}1x10^6$	$3{,}1x10^6$	$2{,}9x10^6$	$2{,}6x10^6$	$2{,}4x10^6$
Nb-97	$9{,}7x10^5$	$3{,}2x10^5$	$1{,}6x10^5$	$1{,}1x10^5$	$8{,}1x10^4$
Rh-103m	$6{,}1x10^8$	$5{,}4x10^8$	$4{,}6x10^8$	$3{,}9x10^8$	$3{,}4x10^8$
Rh-105	$3{,}6x10^7$	$1{,}2x10^7$	$6{,}2x10^6$	$4{,}1x10^6$	$3{,}1x10^6$
Rh-106	$1{,}1x10^7$	$1{,}1x10^7$	$1{,}1x10^7$	1,0x107	$1{,}0x10^7$
Ru-103	$6{,}1x10^8$	$5{,}5x10^8$	$4{,}6x10^8$	$3{,}9x10^8$	$3{,}4x10^8$
Ru-106	$1{,}1x10^7$	$1{,}1x10^7$	$1{,}1x10^7$	$1{,}0x10^7$	$1{,}0x10^7$
Tc-99m	$2{,}8x10^8$	$1{,}1x10^8$	$5{,}7x10^7$	$3{,}8x10^7$	$2{,}8x10^7$
Zr-95	$3{,}0x10^8$	$2{,}8x10^8$	$2{,}5x10^8$	$2{,}3x10^8$	$2{,}1x10^8$

Table 2. Acid waste in a three-batches per week production

Nuclide	1 week (Bq/mL)	3 weeks (Bq/mL)	6 weeks (Bq/mL)	9 weeks (Bq/mL)	12 weeks (Bq/mL)
Ag-111	$1{,}5x10^7$	$9{,}2x10^6$	$5{,}3x10^6$	$3{,}6x10^6$	$2{,}7x10^6$
Ba-137m	$8{,}5x10^6$	$8{,}5x10^6$	$8{,}5x10^6$	$8{,}4x10^6$	$8{,}5x10^6$
Cs-136	$4{,}3x10^6$	$3{,}1x10^6$	$2{,}1x10^6$	$1{,}5x10^6$	$1{,}1x10^6$
Cs-137	$9{,}0x10^6$	$9{,}0x10^6$	$8{,}9x10^6$	8,9x106	$8{,}9x10^6$
I-131	$1{,}3x10^8$	$7{,}7x10^7$	$4{,}5x10^7$	$3{,}1x10^7$	$2{,}3x10^7$
I-132	$1{,}5x10^8$	$6{,}3x10^7$	$3{,}2x10^7$	$2{,}1x10^7$	$1{,}6x10^7$
I-133	$1{,}3x10^7$	$4{,}2x10^6$	$2{,}2x10^6$	$1{,}4x10^6$	$1{,}0x10^6$
Mo-99	$5{,}2x10^8$	$2{,}1x10^8$	$1{,}1x10^8$	$7{,}0x10^7$	$5{,}3x10^7$
Nb-95	$5{,}7x10^7$	$8{,}6x10^7$	$1{,}1x10^8$	$1{,}3x10^8$	$1{,}4x10^8$
Nb-95m	$3{,}0x10^6$	$3{,}1x10^6$	$2{,}9x10^6$	$2{,}6x10^6$	$2{,}4x10^6$
Nb-97	$1{,}9x10^7$	$6{,}4x10^6$	$3{,}2x10^6$	$2{,}1x10^6$	$1{,}6x10^6$
Rh-103m	$6{,}4x10^8$	$5{,}6x0^8$	$4{,}8x10^8$	$4{,}1x10^8$	$3{,}5x10^8$
Rh-105	$1{,}2x10^8$	$4{,}2x10^7$	$2{,}1x10^7$	$1{,}4x10^7$	$1{,}1x10^7$
Rh-106	$1{,}1x10^7$	$1{,}1x10^7$	$1{,}1x10^7$	$1{,}0x10^7$	$1{,}0x10^7$
Ru-103	$6{,}4x10^8$	$5{,}7x10^8$	$4{,}8x10^8$	$4{,}1x10^8$	$3{,}5x10^8$
Ru-106	$1{,}1x10^7$	$1{,}1x10^7$	$1{,}1x10^7$	$1{,}0x10^7$	$1{,}0x10^7$
Tc-99m	$5{,}1x10^8$	$2{,}0x10^8$	$1{,}0x10^8$	$6{,}8x10^7$	$5{,}1x10^7$
Zr-95	$3{,}1x10^8$	$2{,}9x10^8$	2,6x108	$2{,}3x10^8$	$2{,}1x10^8$

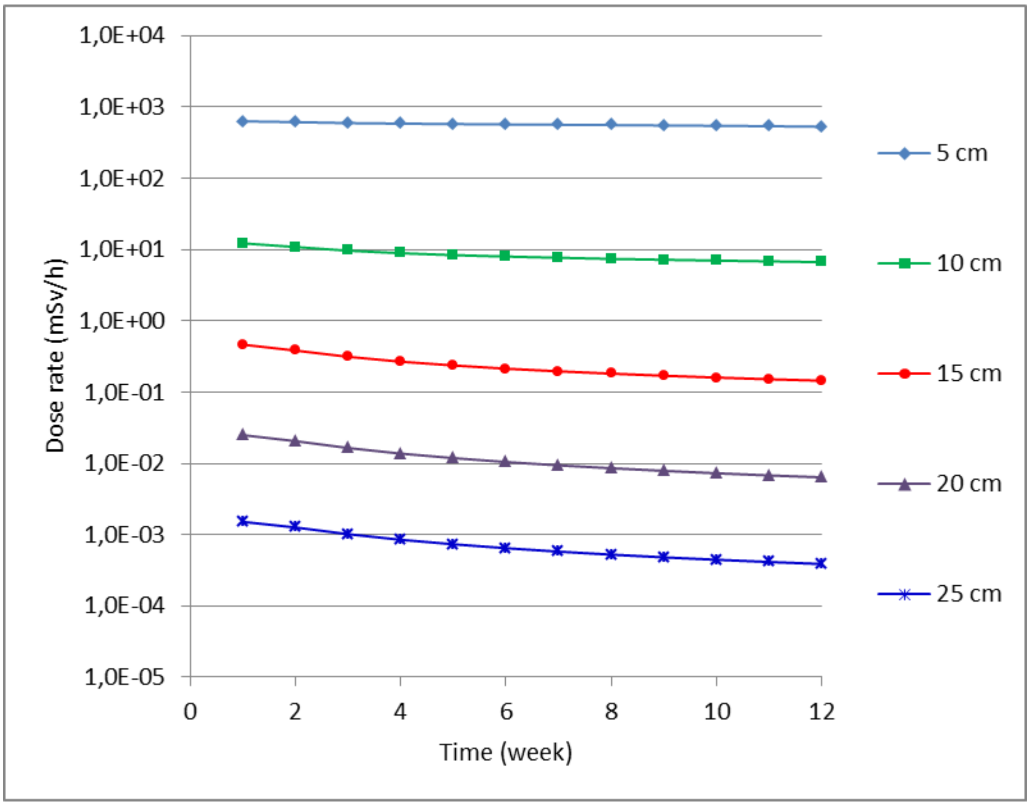

Figure 1. Shielding and dose rate for the acid waste cylinder, 1 batch per week.

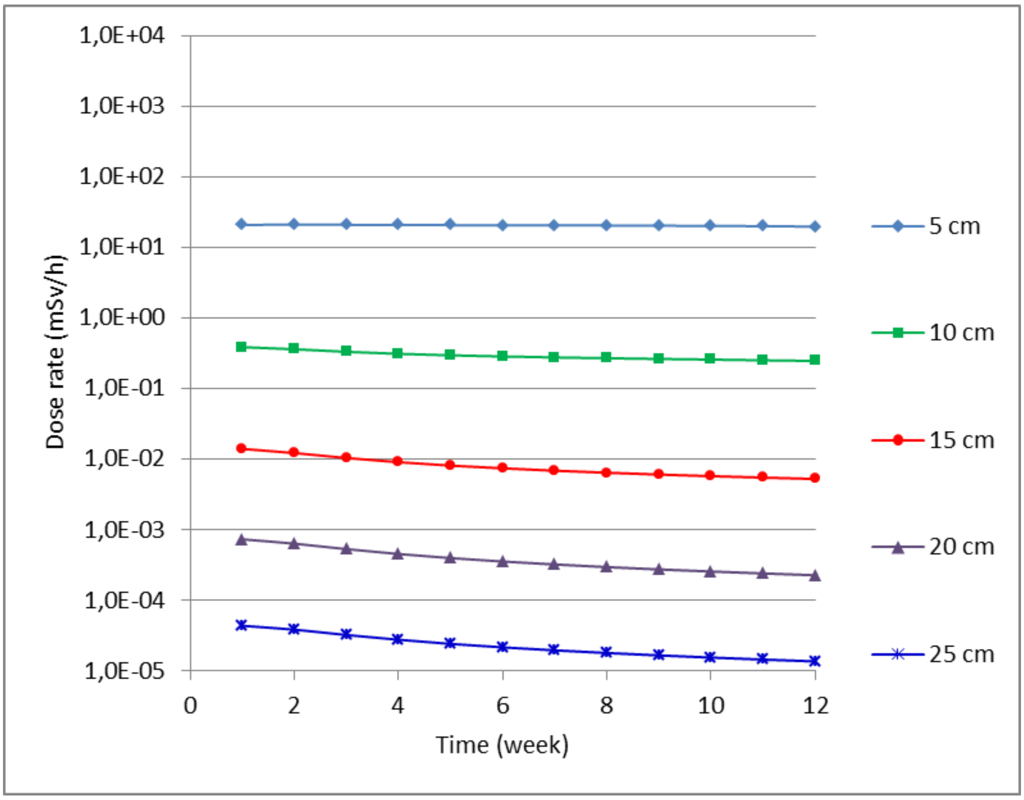

Figure 2. Shielding and dose rate for the alkaline waste cylinder, 1 batch per week.

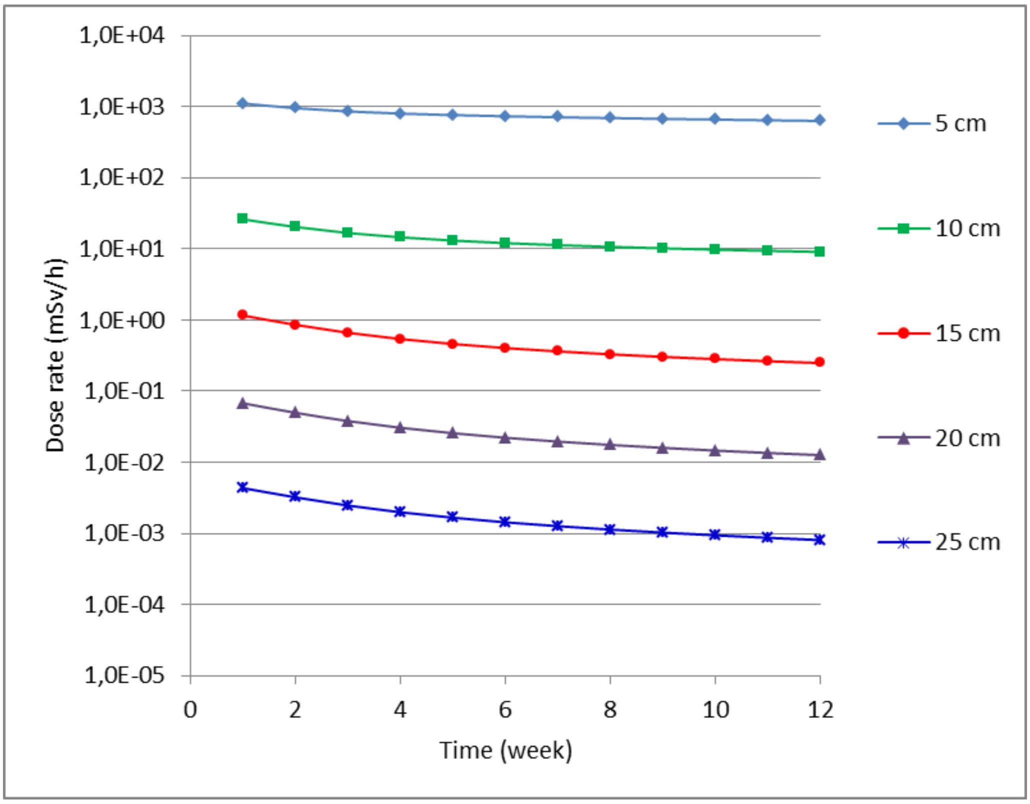

Figure 3. Shielding and dose rate for the acid waste cylinder, 3 batches per week.

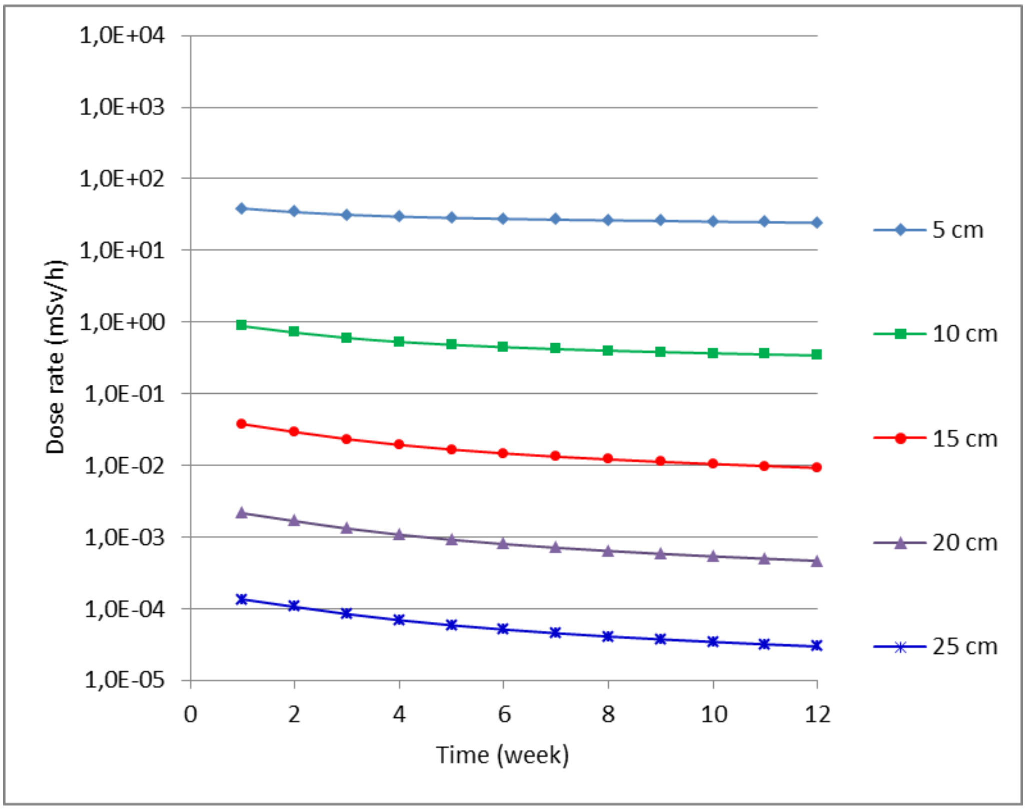

Figure 4. Shielding and dose rate for the alkaline waste cylinder, 3 batches per week.

4. CONCLUSIONS

Results of this study are expected to contribute to the establishment of the radioactive waste management program at the future Brazilian ^{99}Mo production plant. Prediction of the source term, amount of waste to be generated and shielding requirements may help the facility manager to choose the appropriate time for storing the waste inside the ^{99}Mo production facility, before they are transferred to the central waste treatment plant.

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