Study on Shielding Requirements for Radioactive Waste Transportation in a Mo-99 Production Plant – 13382

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ABSTRACT

Brazil is currently planning to produce ^{99}Mo from fission of low enriched uranium (LEU) targets. The planned end of irradiation activity of ^{99}Mo is about 185 TBq (5 kCi) per week to meet the present domestic demand of ^{99m}Tc generators. The radioactive wastes 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 wastes can be predicted based on the yields of fission and activation data for the irradiation conditions, such as composition and mass of uranium targets, irradiation time, neutron flux, production schedule, etc., which were in principle already established by the project management. The transportation of the wastes from the production plant to the treatment facility will be done by means of special shielded packages. An assessment of the shielding required for the packages has been done and the results are presented here, aiming at contributing to the design of the waste management facility for the ^{99}Mo production plant.

INTRODUCTION

Nuclear medicine plays a very important role in medical diagnosis and therapy and presently one of the leading radioisotopes is molybdenum (^{99}Mo) and its daughter technetium (^{99m}Tc), whose current worldwide demand makes many countries take part in a rush for producing it. As Brazilian authorities seek self-sufficiency in radioisotope production, the construction of a facility for ^{99}Mo production has been taken into consideration.

Brazil has already developed its ^{99m}Tc generator[1], IPENTEC, and labeled compound reaction kits for distributing the imported ^{99}Mo throughout the country. The major interest now is the production of ^{99}Mo in order to deliver the required quantity while avoiding dependence on overseas suppliers.

Many methods to produce ^{99}Mo are known but the fission of ^{235}U in low-enriched targets is the one that meets Brazilian needs both in terms of quantity and of specific activity. One drawback of this method is the generation of radioactive wastes with higher activities and longer lived alpha-emitting radioisotopes which require improved management methods. During the target irradiation, many radioisotopes are generated as transuranium elements and fission and activation products. The objective of the present work was to predict the activities that will be handled in the waste treatment facility to be built in the ^{99}Mo production site and calculate the shielding thickness requirements for the packages that will be used to transfer the wastes from the production plant to the waste treatment facility.

The SCALE 6.0[2] software was used to simulate the irradiation of targets, which are made of aluminum-clad uranium-aluminum alloy of approximate composition UAl_2, containing uranium enriched to approximately 20% and calculate the resulting radioisotope inventory of the targets. The thermal neutron flux was set to $1x10^{14}n.cm^{-2}.s^{-1}$ and the irradiation time to 7 days. The reference molybdenum production process considered in this study was alkaline dissolution of the targets, followed by filtration and purification with ion-exchangers.

The shielding of the waste packages was calculated using the MicroShield®-9[3] software. Shielded packages are required for the transportation of the radioactive wastes between the production plant and the waste management facility. The thickness of the shielding must be calculated based on the type, energy and the level of the radiations emitted by the wastes, in order to keep the doses received by the operators within acceptable limits[4].

The results presented in this paper are intended to help the waste management facility designer to optimize shielding and to decide on waste package sizes.

RADIOACTIVE WASTE GENERATION

The reference process adopted in this study produces gaseous, liquid and solid waste streams. Krypton-85, Xenon-133 and a fraction of the iodine isotopes, mainly ^{131}I, are the main gaseous waste products. They are flushed into the *off-gas* system of the hot cell where the dissolution takes place, the noble gases are trapped on special absorbers until they can, by decay, be safely released, while the iodine isotopes are filtered and recovered for use. Solid wastes are generated as spent separation and purification columns in the ^{99}Mo production process as well as other general contaminated disposable materials. However, gaseous and solid wastes will be studied separately and, in this paper, only the results for liquid wastes will be presented.

The reference ^{99}Mo separation process leads to two main streams of radioactive liquid wastes: 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. The activities present in the two liquid waste streams, based on a weekly deliverable production of 1,16E14 Bq of ^{99}Mo, are shown in Tables I and II.

As the volume of the acid stream is expected to be about 10 liters per batch, and as a preliminary choice of the package capacity, the wastes from three batches will be transported to the waste treatment facility. Therefore, the volume considered in Table 1 was 30 liters of acid liquid waste solution. Similarly, the alkaline stream, which is expected to have about 5 liters per batch, will accumulate three batches, thus having a volume of 15 liters to be transported

Table I. Expected accumulated activity of radioisotopes present in the acid liquid waste from three batches of ^{99}Mo production.

Nuclide	Bq/mL	Nuclide	Bq/mL	Nuclide	Bq/mL	Nuclide	Bq/mL
Ag-109m	6.3E+06	Cs-134	5.4E+04	I-133	1.1E+08	Rh-106	1.1E+07
Ag-110	8.1E+00	Cs-134m	1.3E+02	I-135	7.6E+06	Rh-106m	2.1E+02
Ag-110m	5.9E+02	Cs-135	2.1E+01	Mo-99	7.3E+08	Ru-103	6.3E+08
Ag-111	1.5E+07	Cs-136	4.2E+06	Na-24	9.0E+06	Ru-105	2.5E+06
Ag-111m	5.7E+01	Cs-137	9.0E+06	Nb-95	6.0E+07	Ru-106	1.1E+07
Ag-112	5.3E+06	Ge-77	2.5E+05	Nb-95m	2.8E+06	Se-77m	1.6E+04
Ag-113	1.5E+05	Ge-78	9.2E-01	Nb-96	4.9E+04	Se-79	6.6E+00
Ba-137m	8.5E+06	H-3	3.6E+04	Nb-97	3.2E+08	Tc-99	1.1E+03
Br-80	4.6E-01	I-126	1.6E-01	Rb-84	1.3E+01	Tc-99m	4.7E+09
Br-80m	4.3E-01	I-129	6.5E-02	Rb-86	3.8E+04	Zr-93	3.8E+01
Br-82	1.0E+05	I-130	5.4E+03	Rb-88	6.5E+05	Zr-95	3.0E+08
Br-83	1.9E+04	I-131	1.3E+08	Rh-103m	6.3E+08	Zr-97	3.2E+08
Cs-132	2.7E+02	I-132	1.9E+08	Rh-105	3.5E+08		

Table II. Expected accumulated activity of radioisotopes present in the alkaline liquid waste from three batches of ^{99}Mo production.

Nuclide	Bq/mL	Nuclide	Bq/mL	Nuclide	Bq/mL	Nuclide	Bq/mL
Ag-109m	2.6E+05	Cs-134	2.2E+03	I-133	4.6E+06	Rh-106	4.4E+05
Ag-110	3.3E-01	Cs-134m	5.3E+00	I-135	3.1E+05	Rh-106m	8.6E+00
Ag-110m	2.4E+01	Cs-135	8.6E-01	Mo-99	3.0E+07	Ru-103	2.6E+07
Ag-111	6.3E+05	Cs-136	1.7E+05	Na-24	3.7E+05	Ru-105	1.0E+05
Ag-111m	2.3E+00	Cs-137	3.7E+05	Nb-95	2.5E+06	Ru-106	4.4E+05
Ag-112	2.2E+05	Ge-77	1.0E+04	Nb-95m	1.1E+05	Se-77m	6.6E+02
Ag-113	6.0E+03	Ge-78	3.7E-02	Nb-96	2.0E+03	Se-79	2.7E-01
Ba-137m	3.5E+05	H-3	1.4E+03	Nb-97	1.3E+07	Tc-99	4.5E+01
Br-80	1.9E-02	I-126	6.5E-03	Rb-84	5.1E-01	Tc-99m	1.9E+08
Br-80m	1.8E-02	I-129	2.7E-03	Rb-86	1.6E+03	Zr-93	1.6E+00
Br-82	4.2E+03	I-130	2.2E+02	Rb-88	2.6E+04	Zr-95	1.2E+07
Br-83	7.8E+02	I-131	5.1E+06	Rh-103m	2.6E+07	Zr-97	1.3E+07
Cs-132	1.1E+01	I-132	7.7E+06	Rh-105	1.4E+07		

Other liquid wastes generated during the ^{99}Mo are not significant in terms of both volume and activity, consisting mainly in solutions from washing of the purification columns and will be mixed into the main streams previously presented.

PACKAGE SHIELDING

The shielding was calculated using the MicroShield®-9 software, a comprehensive photon/gamma ray shielding and dose assessment package. The inputs are the source geometry, type of shielding material, the radionuclides present in the source and respective activities. The calculation approach was to vary the lead shielding thickness, around a cylindrical vessel with equal diameter and height and the present capacity, aiming at generating graphs of dose rates on the surface and at 1 m from the vessel surface versus shielding thickness. These graphs can be used as reference data sources in the package design, for optimization purposes and compliance with the limits of the Brazilian Regulation for Transport of Radioactive Material CNEN-5.01[5].

The graphs presented in Figs. 1 through 4 show the results of calculations for each liquid waste. The graph in Fig. 1 shows the dose rates delivered by the acid liquid waste with the composition shown in Table I as a function of the lead shielding thickness, calculated at the surface of the package.

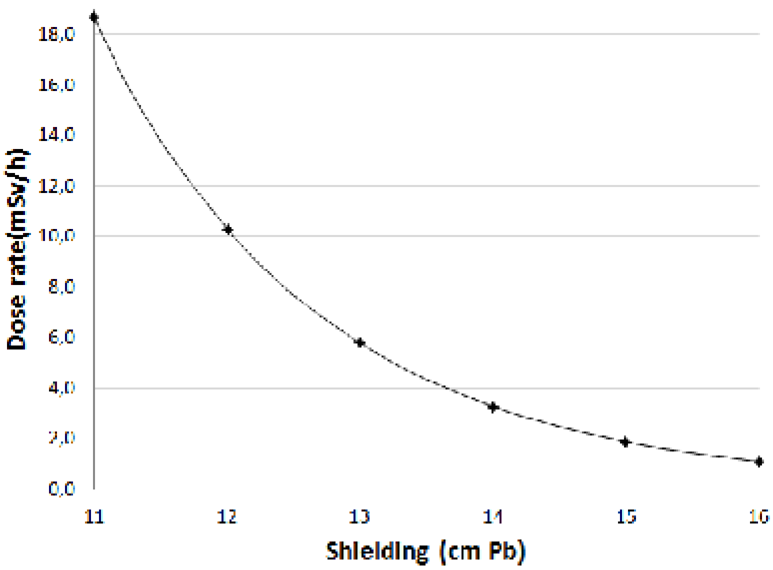

Fig. 1. Dose rates at the surface of the package *versus* the thickness of shielding, for the acid liquid waste stream.

Note that 15 cm of lead are sufficient to meet the dose rate limit of 2 mSv/h, at the surface of the package considered here for transportation of the radioactive waste in a vessel of 30 liters of capacity.

The graph in Fig. 2 shows the results of dose rates calculated one meter from the package surface, indicating that 15 cm of lead will result in a dose rate of 0.26 mSv/h at one meter from the waste package.

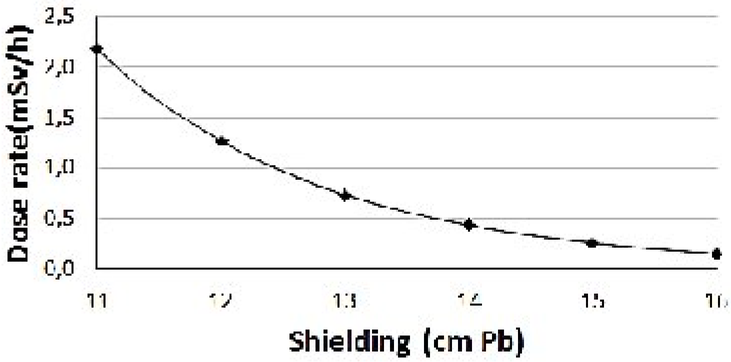

Fig. 2. Dose rates at one meter from the package surface *versus* the thickness of shielding, for the acid liquid waste stream.

The graph in Fig. 3 shows the relationship between the dose rates delivered by the alkaline liquid waste with the radioactive composition shown in Table II and the thickness of lead shielding; these values were calculated at the surface of the package.

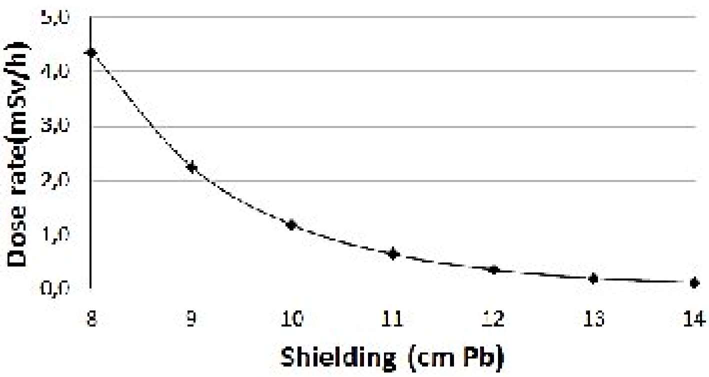

Fig. 3. Dose rates at the surface of the package *versus* the thickness of shielding, for the alkaline liquid waste stream.

For the alkaline waste solution, a shielding of 10 cm of lead is enough to meet the limit of 2 mSv/h criteria, for a vessel with 15 liters capacity.

The graph in Fig. 4 shows the relationship between the dose rate delivered by the alkaline liquid waste, calculated at one meter from the package surface, indicating that a shielding with 10 cm of lead will deliver a dose rate of 0.10 mSv/h, one meter from the waste package.

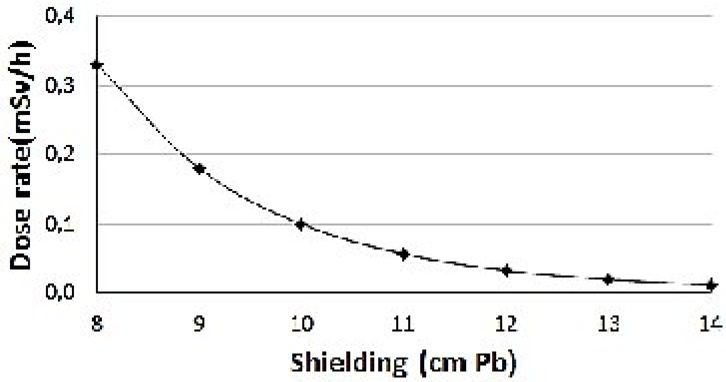

Fig. 4. Dose rates at one meter from the package surface *versus* the thickness of shielding, for the alkaline liquid waste stream.

CONCLUSION

As these packages are to be used only for internal transportation of the wastes, the 2mSv/h dose rate at the surface criteria can be neglected. The package designer can choose different shielding thickness and transportation conditions in order to optimize the radiation protection in the operations to transfer the wastes from the ^{99}Mo production plant to the waste management facility.

These preliminary results of required shielding are in accordance with findings in the literature for similar conditions and when the work is completed it will be useful for optimizing the size of shielding and radiation protection procedures.

FINAL REMARKS

The next step of the present work will be to assess the shielding requirements for vessels with capacities varying for different number of waste batches to be transported. This will allow choosing package sizes, loading equipment, and the frequency of transfers to the treatment facility in order to optimize equipment, operation conditions and the radiation protection in the facility, as required by regulations.

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