Radioactive Waste Issues related to Production of Fission-based Mo-99 by using Low Enriched Uranium (LEU)

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1. Introduction

Molybdenum-99 (Mo-99) is an important radioisotope with a half-life of approximately 66 hrs. It decays to technetium-99m (Tc-99m) which has a life of approximately 6 hrs and this radioisotope is the backbone of modern nuclear diagnostic procedures. Decay scheme of Mo-99 to Tc-99m has been shown in Figure 1.

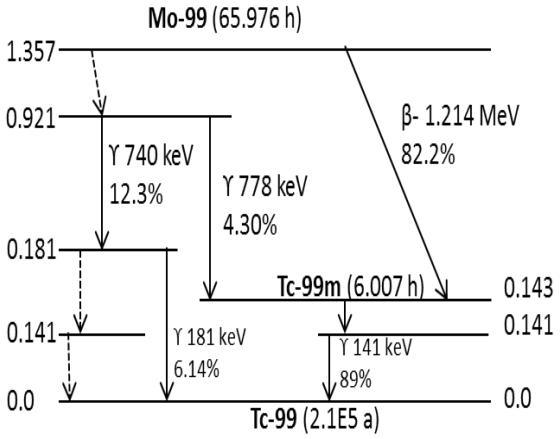

Figure 1. Decay scheme of Mo-99 and Tc-99m

In order to produce fission-based Mo-99 from research reactors, two types of targets are being used and they are highly enriched uranium (HEU) targets with ^{235}U enrichment more than 90wt% of ^{235}U and low enriched uranium (LEU) targets with ^{235}U enrichment less than 20wt% of ^{235}U. It is worth noting that medium enriched uranium *i.e.* 36wt% of ^{235}U as being used in South Africa is also regarded as non-LEU from a nuclear security point of view. In order to cope with the proliferation issues, international nuclear security policy is promoting the use of LEU targets in order to minimize the civilian use of HEU. It is noteworthy that Mo-99 yield of the LEU target is less than 20% of the HEU target, which requires approximately five times more LEU targets to be irradiated and consequently results in increased volume of waste [1].

The waste generated from fission Mo-99 production can be mainly due to: target fabrication, assembling of target, irradiation in reactor and processing of irradiated targets [2]. During the fission of U-235 in a reactor, a large number of radionuclides with different chemical and physical properties are formed [3]. The waste produced from these practices may be a combination of low level waste (LLW) and intermediate level waste (ILW) comprised of all three types, i.e., solid, liquid and gas. Handling and treatment of the generated waste are dependent on its form and activity. In case of the large production facility, waste storage facility should be constructed in order to limit the radiation exposures of the workers and the environment.

In this study, we discuss and compare mainly the radioactive waste generated by alkaline digestion of both HEU and LEU targets to assist in planning and deciding the choice of the technology with better arrangements for proper handling and disposal of generated waste.

2. Summary of Rad-waste Issues

Irradiated targets are loaded into the hot cell where the target plates are inserted into the dissolver to separate the Mo-99 from other fission products.

During the dissolution process, a number of volatile and radioactive gases are produced. The main active gases produced in the dissolver both for HEU & LEU targets during the alkaline dissolution are given in the Table 1.

Table 1. Major Off-gases Produced during Dissolution

Fission Product	Half Life
Xe-131m	11.93 d
Xe-133m	2.19 d
Xe-133	5.24 d
Xe-135	9.14 h
I-131	8.02 d
Kr-85	4.48 h

Activity and volume of the produced gases depends on the cooling time of the targets. In current practices, the off-gases are stored in especially designed/shielded delay/decay cascades and are released into the environment after decay as required by the applicable regulations. The off-gases handling system of the facility should be efficient to hold certain volumes of these fission gases. The amount of the fission gases to be released into the environment is based on the regulatory limits set by the licensing authority/regulatory body (e.g., in NUREG-0472, the condition on the normal release of Xe-133 at the

boundary of the facility dose rate limits should not exceed 500 mrem/year for the total body and 3000 mrem/year for skin). Stringent monitoring arrangements are required in order to avoid any leakage of these gases into the processing area and environment.

Production of radio xenon during the dissolution process is of major concern as radio xenon is a signature gas of any nuclear test and its release/leakage from the isotope production facility may generate spurious signals for international monitoring system (IMS). In order to control this situation, CTBTO has recommended the release limit for radio xenon releases as less 5 GBq/day from radioisotope facilities. Different studies have also been carried out and found that a maximum release of 5 GBq/day can be a safe limit and has no effect on IMS [4]. Moreover, production of hydrogen during alkaline digestion of target plates can be a safety concern. The accumulation/leakage of hydrogen may cause explosion and may be a source of release of fission product gases into the process area.

In both type of the dissolution processes, i.e., alkaline and acidic, liquids are being used in the dissolver in order to dissolve the irradiated targets. In the production of Mo-99, main stream of liquid waste arises from the dissolver and filtration process. There are two possible streams of liquid waste, i.e., LLW & ILW. Based on the technical reasons, these two types of wastes should be kept separately. Inappropriate design of the storage tanks may cause leakages of the liquid waste.

The generation of liquid waste is different for both types of fuel targets used for fission based Mo-99 production. In order to produce the same amount of Mo-99 from the LEU target, it must contain uranium ≥5 times of the uranium in the HEU target. As a result, 3 to 6 times more dissolver solution will be required. This will increase the liquid waste as well as increase the quantity of solid waste [3].

Table 2. Liquid waste generated in Mallinckrodt/ECN, Netherlands (93% enriched HEU UAl_x plate targets, 3000 Ci) [5]

Waste Form	Volume in (liter)	Radioisotopes	Activity (MBq/l)
ILW	18.7	^{89}Sr	740
		^{90}Sr	630
		^{137}Cs	6400
		^{103}Ru	500
		^{106}Ru	46
		^{125}Sb	4.6
LLW	12	^{103}Ru	5.040
		^{106}Ru	0.4637
		^{125}Sb	0.046

The main source of solid waste in Mo-99 production process mainly consists of filtered uranium and TRU, the ion exchange resins and absorber columns. It may consist of charcoal filters, different hot cell filters, valves, pumps, tubes, etc. According to IAEA-TECDOC-1051, Management of Radioactive Waste from Mo-99 Production, about 26 times more Pu-239 is produced in LEU targets than HEU targets with comparable Mo-99 yield which increase the disposal cost significantly.

3. Conclusions

With the use of the LEU targets in Mo-99 production facility, significant increase in liquid and solid waste has been expected. Moreover, in order to produce same quantities of Mo-99 as from the HEU, more targets of the LEU are required to be irradiated and processed which may increase the reactor operation time as well as processing facility operation time. Therefore, it may be necessary to pay more attention to the safety issues related to the radioactive waste management in the production of Mo-99 via LEU targets.

Acknowledgments
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