# RADIOACTIVE WASTE RESULTED IN $^{99}$MO FISSION PRODUCTION

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## ABSTRACT

Management of radioactive waste is an integral and very important part of $^{99}$Mo production, especially for fission $^{99}$Mo, and should be given high priority during production process development and operation. To ensure a continued reliable and cost-effective supply of $^{99}$Mo, the management of associated waste must be carried out in accordance with internationally accepted safety criteria and related national regulations.
The waste products form and characteristics will be dependent on the process design and chemistry. Typically, vital aspect of the Fission Production Mo processing is the holding of waste streams at the site of production for initial decay of the shorter lived fission products. This usually follows a sequence of in-cell storage, storage adjacent to the hot cell operations, and transfer to longer term storage or processing. Adequate capacity must be allowed for this initial waste storage at the hot cell area

**Key words: molybdenum-99, fission molybdenum, molybdenum waste**

## Introduction

Molybdenum-99 ($^{99}$Mo) and its daughter product $^{99m}$Tc are the most important radionuclides used in nuclear medicine practice. The importance of these radionuclides stems from the fact that $^{99m}$Tc is used in about 80% of all diagnostic nuclear medicine procedures. $^{99}$Mo production for $^{99m}$Tc generators gives rise to a number of waste streams, some of which need special care in handling. Radioactive waste from the production of $^{99}$Mo must be managed in such a way that the protection of people and the environment is ensured, both now and in the future.

$^{99}$Mo is produced mainly by nuclear fission of $^{235}$U. Processing of irradiated uranium targets for $^{99}$Mo production generates considerable amounts of radioactive wastes containing fission products as well as small quantities of transuranium elements. Small amounts of Mo are also produced by neutron activation of $^{98}$Mo targets. However, this method is not yet fully developed for medium or large scale production.

The nuclear and radioactive waste management is based on well-established safety standards for the management of radioactive waste. Romanian legislation and regulations are all developed from internationally agreed standards, guidelines and recommendations in order to ensure the protection of public and the environment.

Molybdenum production by separating out of the fission products resulted from the fission of uranium involves four stages [1].

First stage is the *manufacturing of the target* to be irradiated. The target used in the acid dissolution method is a uranium foil, and for the alkaline process is a uranium plate.

The second stage is the *irradiation of the target* in the reactor; after a cooling period the irradiated target is transferred to a post irradiation facility for further processing.

The third stage involves the *chemical processing of the target*. This stage implies the dissolution of the target in acid or alkaline solution. The waste resulted depends on the process.

Lastly, the fourth stage deals with the *management of the radioactive waste*. All the materials used for the separation of $^{99}$Mo become radioactive waste. Although some of these materials are high level waste (HLW), most of them are intermediate level waste (ILW).

## Waste characteristics

Production of $^{99}$Mo by means of neutron induced fission of $^{235}$U (as HEU or LEU) is operated in some countries on an industrial scale. Although this production method supplies nearly the whole worldwide demand for $^{99}$Mo, it also generates the most radioactive waste of all production methods [2]. This arises from the fact that a wide range of nuclides are formed during $^{235}$U fission.

The waste products form and characteristics will be dependent on the process design and chemistry. The waste streams to be handled are summarized below:
• *Solids* from processing, such as precipitates or cans that held targets.
• *Liquids* from processing are common to all Mo-99 production processes. These are usually intermediate level waste at time of production and often remain so for long time periods due to the fission product content. There is more than one stream of liquid waste from target processing and Mo-99 purification and these may be of similar or differing chemistry. These streams will contain different major nuclides or different concentrations of the same nuclides.
• *General cell solid* waste must be considered and incorporated into the “existing” ILSW (Intermediate Level Solid Waste) handling system. In-cell consumables such as tubing, columns and wipes, fittings, etc, are regularly replaced and are accumulated for batch-wise removal and shielded transfer to a shielded storage facility.
• *Gaseous* waste arises from all forms of processing and must be handled effectively for safety and regulatory reasons. For FPMo (Fission Product Mo-99) the iodine and the noble gases in the streams must be held for sufficient decay or treated before gases can be released to the extract system and stack.
• *Low level solid* waste will be produced by hot cell processing (at the rear of cell operations and transfers, and for any decontamination activities). This must be incorporated in the existing site low level operating system.
• *Low level liquid* waste is produced from decontamination and other activities at rear of cells and also must be planned for, and incorporated into the existing site system by either a batch transfer system or a reticulation system.

Vital aspect of the FPMo processing is the holding of waste streams at the site of production for initial decay of the shorter lived fission products. This usually follows a sequence of in-cell storage, storage adjacent to the hot cell operations, and transfer to longer term storage or processing. Adequate capacity must be allowed for this initial waste storage at the hot cell area.

Other aspects to consider for the FPMo route are Safeguards and the tracking and security of the uranium throughout the process. The uranium deports to different waste product streams depending on whether acid processing or alkaline processing is used for target dissolution.

## FPMo acid processing route

The acid route in obtaining FPMo uses a LEU foil target. The irradiated target is dissolved in nitric acid. During the dissolution of the LEU foil fission gasses the rest of the gasses resulted (NOx) are drained from the dissolver using a cold finger gas trap. By this method the gasses are treated as solid waste.
On further are presented the wastes resulted during the acid processing route in obtaining FPMo:
• *Liquid waste*. A primary liquid waste results from the dissolution of the targets in nitric acid (or other acid mixtures) after the extraction of the Mo-99. Due to the total dissolution of the target this results in the major proportion of the mixed fission products reporting to the liquid waste. This is a longer lived waste, dominated over the long term by Cs-137 and Sr-90. In the medium term Ce-144, Ru-106/Rh-106, Zr-95, Nb-95 also contribute significantly to the activity. This is accumulated in cell on a filling and hold for decay basis before removal from the cell. This waste will required further storage, either at location or in a separate area and may be combined in larger tanks, subject to criticality assessment and certification.
• *Liquid waste*. A secondary liquid waste results from the purification steps on the original Mo-99 adsorption column. This may be similar in chemistry to the primary solution but more dilute in most species. This is similarly dominated over the longer term by the Cs-137 and Sr-90.
• *Solid waste* from target cladding or enclosure. This may be aluminum or stainless steel and of high initial activity and are accumulated in cell until a combined batch is removed as intermediate level solid waste.
• *Solid waste* from hot cell operations. This includes other cell waste, or consumables, such as adsorption columns, tubing and other redundant equipment. This is also accumulated in cell and removed on a campaign basis with the other ILSW. Both these intermediate level solid wastes are contaminated with mixed fission products and must be transferred in shielded containers to shielded storage.
• *Other in cell liquid wastes* that are produced in smaller volume. These have been accumulated at one operation, transferred separately in a shielded flask and accumulated in a storage facility. Due to the lower activity and shorter lived nuclide content this waste can be processed and disposed of through the low level liquid waste treatment system after a moderate storage time.
• *Gaseous waste* is generated during the target dissolution and must be treated and/or stored before HEPA and charcoal filtration before passing to the stack within the regulatory requirements. This does lead to other wastes which periodically have to be removed, stored and processed. The fission gas traps (treated charcoal) the HEPA filters and charcoal filters all contribute to waste.
• *Low level solid waste* from rear of cells activities (decontamination from items or transfer operations).
• *Low level liquid waste* from rear of cells decontamination activities. Since this liquid waste will contain mixed fission products it may or may not be suitable for direct treatment in the effluent plant. A pretreatment may be required.

Waste Facilities required at FPMo Processing Plant that uses the acid processing route.
• Heavily shielded Hot Cells for processing targets and initial storage of solid and liquid wastes. Hot cells fitted with master-slave manipulators and with extract ventilation through HEPA and charcoal filtration. Cells to have means of transferring waste material to shielded flasks or containers, or shielded transfer mechanism to storage facilities adjacent or near to the hot cells.
• Overhead crane and/or other mechanism to move and accurately locate heavily-shielded flasks to allow transfers of waste.
• In cell storage tanks for accumulation of liquid wastes (possibly with segregation of types of liquid waste).To have sufficient capacity to hold waste for sufficient time to allow significant decay of shorter lived nuclides.
• In cell storage ability to hold solid waste for removal on a (full) batch basis.

• Additional storage and/or processing capacity to transform liquid waste into a solid product in shielded facilities suitable for long term storage or disposal.
• Safeguards monitoring of this waste since this contains the uranium from the targets.

## FPMo Alkaline Processing Route

The alkaline processing route uses a LEUAlx plate target. The general waste regimes are as follows:
• *Uranium precipitate*. For alkaline dissolution the uranium remains as a precipitate in a filter at the base of the dissolver. This dissolution technique results in a distribution of the fission products between the solution and the precipitate. The filters containing the filter cake are removed from the dissolver. There are some variations on locally dealing with this material from capping and interim storage in cell, from removal and transfer to a different vessel, to redissolving and reprecipitating in a different filter for subsequent encapsulation. No producer world wide is currently reprocessing this precipitate for uranium recovery whereas several producers are multiple encapsulating (welded sealing) for longer interim storage in shielded facilities. The longer lived isotope Sr-90 reports to the uranium precipitate.
• *Primary intermediate level liquid waste*. This waste results from the dissolution liquid after removal of the Mo-99. The liquid contains the Cs-137 and therefore remains as intermediate level. Due to the short lived nuclides also present it is necessary to store this in-cell or beneath cell (underground) to allow sufficient decay before shielded transfer (or transport) to further consolidated storage or processing to a solid product.
• *Secondary level liquid waste*. This may be alkaline or can be acid depending on the Mo-99 purification flowsheet. Often it may be a number of streams which are combined for storage. This liquid waste will initially be classified as intermediate level but has potential to decay to the low level category in a reasonable period (years). Similarly, this waste is accumulated at the site of production to allow initial decay followed by transfer to accumulated storage for further decay and processing to solid.
• *Solid cell waste* which is collected in cell(s) and removed by the shielded ILSW transfer system to the shielded ILSW storage system. This waste includes the columns, tubing, fittings and other general cell waste.
• *Gaseous waste*. There is a requirement to retain the off gas from the dissolution process to allow for sufficient decay of the noble gases before release through the cell extraction HEPA filtration and carbon adsorption systems.
• *Low level solid waste* from rear of cells activities (as acid route)
• *Low level liquid waste* from rear of cells decontamination activities. Since this liquid waste will contain mixed fission products it may or may not be suitable for direct treatment in the effluent plant. A pretreatment may be required.

Waste Facilities required at FPMo Processing Plant that uses the alkaline processing route.
• Heavily shielded Hot cells for processing targets and initial storage of solid and liquid wastes. Hot cells fitted with master-slave manipulators and with extract ventilation through HEPA and charcoal filtration. Cells have to have means of transferring waste material to shielded flasks or containers adjacent or near to the hot cells.
• Overhead crane and/or other mechanism to move and accurately locate heavily-shielded flasks to allow transfers of waste.
• In cell storage tanks for liquid wastes (with segregation of types of liquid waste).To have sufficient capacity to hold waste for sufficient time to allow significant decay of shorter lived nuclides.
• In cell storage to hold uranium precipitate in suitable containment to allow decay of shorter lived nuclides and ability to transfer material to further processing (encapsulation) without spread of contamination. Safeguards monitoring of this waste since this contains the uranium from the targets.
• In cell storage ability to hold general solid waste for removal on a (full) batch basis.
• Additional storage and/or processing capacity to transform liquid waste into a solid product in shielded facilities suitable for long term storage or disposal.

## Conclusion

Radioactive waste from the production of Mo, which is the most important radionuclide for medical application, must be managed in such a way that the protection of people and the environment is ensured, now and in the future.

The volumes and types of radioactive wastes generated by $^{99}Mo$ production are largely dependent on the production method. Most methods used for $^{99}Mo$ production are based on nuclear fission of targets containing $^{235}U$. Processing of these targets generates considerable amounts of radioactive wastes containing long lived fission products and α-emitters. The presence of fission products and actinides in solid and liquid radioactive wastes complicates all stages of waste management. The treatment and conditioning of fission $^{99}Mo$ production wastes require more progressive technology and equipment, hot cells remote handling, shielded waste packages, and capacity for interim storage. The waste will be generated as solids, liquids, and/or gases, and will include material in the low, intermediate and even high level radioactive categories. Initial treatment of waste streams is usually required at the production site, prior to short or long term storage. The treatment required is dictated by both the form of the waste and its activity level. Final disposal of waste should be considered right from the beginning of process development.

## References

[1] Laurentiu c. Dinu, Stefan d. Dulugeac and Ana Maria Ivan - Time evolution for the activity of radioactive waste associated to $^{99}Mo$ production using LEU targets – Journal of Nuclear Research and Development, no.6 Dec. 2013;

[2] IAEA VIENNA IAEA-TECDOC-1051 – Management of radioactive waste from $^{99}Mo$ production