RADIOACTIVE WASTE TREATMENT FROM MO-99 PRODUCTION FACILITY IN THE NETHERLANDS

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ABSTRACT

In Petten, the Netherlands, a Molybdenum Production Facility (MPF) has been realised since 1994 by the Nuclear Research and Consultancy Group (NRG) in close co-operation with Mallinckrodt, a company at the same site that produces isotopes for medical applications. The facility is operated by Mallinckrodt personnel under the license of NRG. At this moment the facility has a weekly production rate of 0,4 PBq. (10,500 Ci). COVRA is the Central Organisation for Radioactive Waste in the Netherlands and responsible for the execution of the radwaste policy in the Netherlands.

The management of the radioactive waste treatment, developed by Mallinckrodt, NRG and COVRA, has been one of the key issues during the development of the production facility.

For solid and liquid low level waste (LLW) an existing route at COVRA was available.

For the handling of solid and liquid intermediate level waste (ILW) from the MPF, a new system for transport, interim storage and treatment had to be developed.

The waste management project included the following issues:

- development, construction and operation of a filling and packaging station for liquid waste at the production site.
- design and realisation of interim storage and treatment facilities for both the solid and liquid ILW at the COVRA site.

In 2000 the completed facilities have successfully undergone the hot testing phase.

The experiences achieved confirmed the integration of the waste management in the molybdenum production cycle. This paper will give an overview of the project and the actual results of the waste treatment operations.

INTRODUCTION

In nuclear medicine practice molybdenum (^{99}Mo) and its decay product technetium (^{99m}Tc) are very important radionuclides. With a world-wide growing demand of these nuclides a new Molybdenum Production Facility (MPF) was planned in the Netherlands.

The MPF has been realised in Petten since 1994 by the Nuclear Research and Consultancy Group (NRG) in close co-operation with Mallinckrodt, a company at the same site that produces isotopes for medical applications.

The Molybdenum is produced by the method of nuclear fission of Uranium-235. With this production method the generation of a variety of radioactive waste was recognised as a key issue, that should be given high priority during the development of the production process, the operational and the post-operational phase. The management scheme for radioactive waste was jointly developed by Mallinckrodt, NRG and COVRA, the Central Organisation for Radioactive Waste in the Netherlands. **(ref. 1)**

Mo PRODUCTION METHOD

Target plates consist of an aluminium-cladded uranium/aluminium alloy of the approximate composition UAl_3. To achieve the highest possible fission product yield, uranium, enriched to 90-93% (HEU), is irradiated in the HFR-reactor at Petten with a thermal neutron flux of $1x10^{14}$ $n.cm^{-2}.s^{-1}$. The irradiated targets are transported to the Molybdenum Production Facility (MPF) where the targets are dissolved in an alkaline liquid, followed by chemical treatment steps passing several absorber columns and ion-exchangers.

WASTE GENERATION

The volatile fission products with xenon (^{133}Xe) as the main isotope are flushed by a stream of nitrogen. They leave the dissolver at its upper end and pass through a reflux condenser, which condenses the water resulting from the oxidized hydrogen generated during the digestion process.

The separation process of the HEU-target leads to an alkaline waste stream arising from the dissolution of the targets in sodium hydroxide and an acidic waste stream arising from passing through ion-exchangers. The alkaline waste solution contains nearly the total amount of aluminium from the target and an essential amount of fission products. The characteristics of the main liquid streams are compiled in table 1. The acidic waste stream consists of sulphuric acid containing thiocyanate, sulphite and the remaining fission products.

Table I: Actual characteristics of liquid waste per hot cell after production of 3000 Ci ^{99}Mo

Liquid from	**Volume (l)**	**Content (g/l)**		**Activity (Mbq/l)**	
Cell 1 ILW	8,2	NaOH Al U	240 20 0,005	^{89}Sr ^{90}Sr ^{137}Cs	740 630 6400
Cell 2 ILW	10,5	$NaNO_3$ NaOH	102 29,6	^{103}Ru ^{106}Ru ^{125}Sb	500 46 4,6
Cell 3 LLW	7,3	Na_2SO_4 NaI Na_2SO_3 NaOH NaSCN	115 0,06 121 16 4	^{103}Ru ^{106}Ru ^{125}Sb	5 0,46 0,046
Cell 4 and 5 LLW	4,7	NaOH $NaNO_3$	12 17	^{103}Ru ^{106}Ru	0,040 0,0037

The Mo purification process generates only small volumes of liquid waste with relatively low activity level and consist mainly of aqueous solutions of low concentration of acidic or alkaline media.

Solid waste is generated mainly from spent ion exchange resins and absorber columns. Other types of solid waste are filter material from off-gas cleaning, such as activated charcoal filters or absolute filters from the hot cell. Small amounts of solid waste arise from the replacement of parts of the whole Mo production plant like valves, pumps, tubes, etc. The characteristics of the main solid waste stream are compiled in table 2.

Table IIActual estimated characteristics of solid waste per hot cell after production of 3000 Ci ^{99}Mo

Solid Waste, type	**Components**	**Activity some days after production (MBq/syntacscontainer)**
Cell 1, ILW	filters sinter metals tissues pipes/tubes	^{234}U 0,93 ^{235}U 0,029 ^{236}U 0,0037 ^{239}Pu 0,011 ^{89}Sr 140 000 ^{90}Y 500 ^{91}Y 150 000 ^{95}Zr 150 000 ^{95}Nb 10 000 ^{95}Zr 150 000 ^{103}Ru 120 000 ^{106}Ru 1 600 ^{141}Ce 290 000 ^{144}Ce 31 000 ^{125}Sb 17 To be transported in type A container after two months decay-time
	Absorber columns Ion exchangers PVC tubes stainless steel components tissues	^{131}I 23000 ^{103}Ru 420000 ^{106}Ru 23000
Cell 3, ILW	Absorber columns polyethylene bottles	^{103}Ru 30000 ^{106}Ru 3300
Cell 4, LLW	tissues	Mo-99 tracers
Cell 5, LLW	tissues	Mo-99 tracers
Replacement components when necessary, ILW/LLW	dissolvers drop collector in cell storage tanks	various activity

The remaining U and fission products are retained in the dissolver by Bio-Rex-5 containing the highest activity such as lanthanides, ruthenium, zirconium, etc. The material of the spent target is sent back to the target producer for reprocessing. There will be just some traces of U and α-emitters in the liquid waste.

RADIOACTIVE WASTE MANAGEMENT

During the planning, design, construction and operational phase of the MPF the implementation of the waste management concept has played an important role. Already during the licensing procedure, the strategy for waste management including the route for the targets and long lived fission products had to be described and was approved by the responsible authorities. Other important issues such as criticality of U and decay heat generated by the radionuclides were taken into consideration.

Waste is generated as solids, liquids and gases and include low (LLW), intermediate (ILW) and short-lived high level wastes (HLW). For solid and liquid LLW an existing treatment route at COVRA was available. For the handling of solid and liquid ILW from the MPF, a new system for transport, interim storage and treatment had to be developed. Therefore the Mo-waste project was established in 1994.

This project included the following issues:

- development, construction and operation of a filling and packaging station for liquid waste at the production site,
- design, engineering, fabrication and testing of Type A-transport containers for both the solid and liquid ILW,
- modification of transport vehicle,
- design and realisation of interim storage and treatment facilities for both the solid and liquid ILW at the COVRA site.

WASTE-TREATMENT AT THE PRODUCTION FACILITY

Gaseous waste

Hydrogen is oxidized to H_2O via CuO at 350-400°C and the resulting water is condensed together with tritium (^3H). Xenon is collected together with nitrogen in a pre-evacuated stainless steel tank. The gas is then sent to the Xenon delay system, which consists of carbon filters. To guarantee a safe, leakage-free process all equipment used in this extremely hot part of the process is made of stainless steel.

Liquid waste

To avoid Al-precipitation the alkaline solution is adjusted with 6M NaOH and limited to 40 g/l Al. For decay of iodine (^{131}I) and other short lived fission products the alkaline solution is stored in stainless steel tanks for about 12 months. The acidic waste stream consists of 2M sulphuric acid and the remaining fission products.

After this storage period the liquids are pumped with the filling and packaging station into specially designed liquid holders of 44 litres. These are filled until the activity reaches the IAEA Type A-2 transportrequirements. The liquid holders are placed in a shielded type A transport container for liquids for shipment to COVRA.

Other low level liquid waste streams are stored in buffer tanks for a period of several months. Thereafter the liquid is pumped in standardised 30 litres liquid waste drums and transported unshielded to COVRA.

Solid waste

The waste is collected within the hot cells and loaded into small waste boxes and racks. Spent columns are stacked for a decay-period of several months within the cell. Other waste is put in so called syntacs-containers with a diameter of 210 mm and a length of 650 mm. For replacement of bigger components a syntacs-container of 295/690 is available. These containers with waste are, depending of the dose-rate on surface, packed after pre-compaction in standard 100-drums for the super-compaction route of COVRA or packed in shielded type A transport containers for shipment to COVRA. A flowscheme with the waste process steps are presented in figure 1.

The material of the spent target is sent back to the producer of the targets. It is possible that in the future waste from the reprocessing will be sent back to the user.

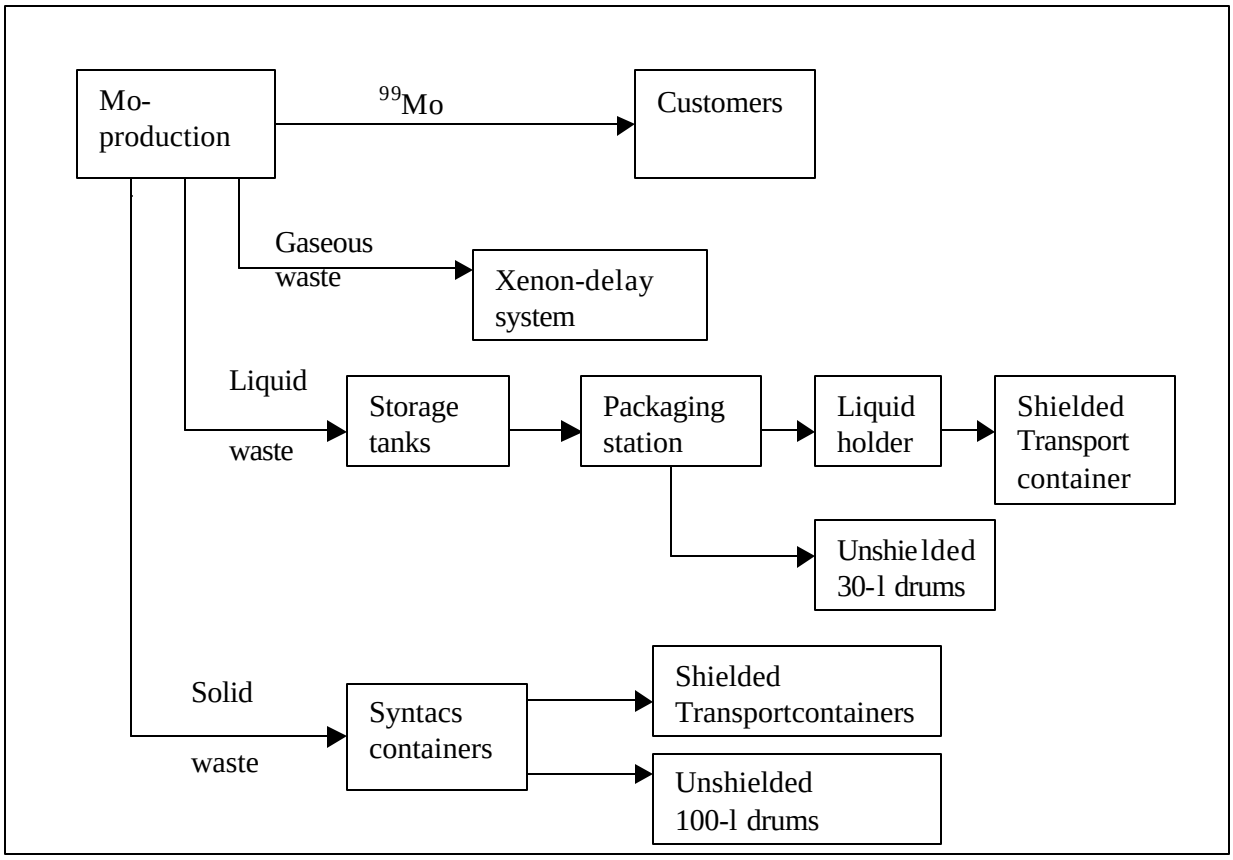

Fig.1. Waste process steps at the Mo-production facility

WASTE TREATMENT, CONDITIONING AND LONG-TERM INTERIM STORAGE FOR 100 YEARS AT COVRA

Shipment of the waste from NRG/Mallinckrodt site to the COVRA site takes place by road using a modified COVRA-owned truck. With these transports the IAEA transport-rules are taken into account. The first transport with solid waste took place in 1996 and the first liquid waste transportation in 1997.

Liquid waste

The low level liquid wastes are treated at COVRA together with other LLW liquids from hospitals and laboratories. The activity is concentrated by means of chemical coagulation and co-precipitation. The precipitates are separated and conditioned with cement. After control of the activity, the purified water is discharged into a nearby river. The liquids with the higher activity are first handled in a dedicated reception-installation for emptying the liquid holder by bore-drilling them from top to bottom. The liquid holder is packed in a standard 100-l drum for the supercompaction route. The liquid is then stored in 2 stainless steel buffer tanks of 3000 litres awaiting in-drum immobilisation in 200-l drums with a mobile cementation unit. The maximum activity within the waste packages comply to the A-2 transport requirements. The 200-l drums are placed and fixed in a 1000-l concrete overpack.

Solid waste

For the treatment of the syntacscontainers the following routes are available. Syntacs-containers with a surface dose rate above 10 mSv/h are stored in a shielded interim storage bunker for a decay period of approximately two years. Depending on the dose rate of the syntacs containers after two years of storage the containers will be placed and embedded in cement directly in 200-l drums or follow the supercompaction route.

Syntacs-containers with a dose rate below 10 mSv/h on the surface will be prepressed and put in a standard 100-l drum for solid waste. These drums follow the route for supercompaction and the resulting parcels are embedded with cement in 200-l drums. The conditioned waste packages are placed in an interim storage building for a period of 100 years.

CONCLUSIONS

- During the development and commissioning phase of a Molybdenum Production Facility, radioactive waste management is one of the key issues.
- With the realisation of the new waste treatment facilities at the site of the Mo producer and at the site of the waste operator, proper care is taken for the radioactive waste management aspects of Mo-production in the Netherlands.

REFERENCES

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